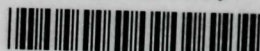


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ENVIRONMENTAL MONITORING AT
ARGONNE NATIONAL LABORATORY

ANNUAL REPORT FOR 1975

by

N. W. Golchert, T. L. Duffy,
and J. Sedlet

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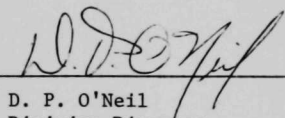

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ENVIRONMENTAL MONITORING AT ARGONNE NATIONAL LABORATORY

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ABSTRACT

The results of the environmental monitoring program at Argonne National Laboratory for 1975 are presented and discussed. To evaluate the effect of Argonne operations on the environment, measurements were made for a variety of radionuclides in air, surface water, Argonne effluent water, soil, grass, benthos, and foodstuffs; for a variety of chemical constituents in surface and Argonne effluent water; and of the environmental penetrating radiation dose. Sample collections and measurements were made at the site boundary and off the Argonne site for comparison purposes. Some on-site measurements were made to aid in the interpretation of the boundary and off-site data. The results of the program are interpreted in terms of the sources and origin of the radioactive and chemical substances (natural, fallout, Argonne, and other) and are compared with accepted environmental quality standards. The potential radiation dose to off-site population groups is also estimated.

I. INTRODUCTION

A. General

This report is prepared to provide the U. S. Energy Research and Development Administration (ERDA) and the public with information on the level of radioactive and non-radioactive pollutants in the environment of Argonne National Laboratory and on the amounts of pollutants, if any, added to the environment as a result of Argonne operations. The report follows the guidelines given in ERDA Manual Chapter 0513.⁽¹⁾ The Laboratory conducts a continuous environmental monitoring program on and near the Argonne site whose primary purpose is to determine the magnitude, origin, and identity of radioactive or potentially toxic chemical substances present in the environment. Of special interest is the detection of any such material released to

the environment by Argonne. One important function of the program is to verify the adequacy of Argonne's effluent pollution controls.

Argonne is a multi-disciplinary research and development laboratory with several principal objectives. It carries out a broad program of research activities in the physical, biomedical, and environmental sciences and serves as an important center for energy research and development, both nuclear and non-nuclear. Some of the energy-related research projects are a major effort in the fast breeder nuclear reactor (liquid-metal and gas-cooled), reactor safety studies, improvements in the utilization of coal for power production, the development of electric batteries for vehicles and off-peak energy storage, magnetohydrodynamic power generation, and solar energy utilization. Environmental research studies include a Great Lakes radioecology program, which is primarily concerned with the effects of effluents from nuclear and fossil power plants on Lake Michigan and other watersheds, studies on the dispersion and behavior of airborne pollutants under various meteorological conditions, and reclamation of strip-mined lands. Almost all of the work at the Laboratory is of an unclassified nature.

The principal nuclear facilities at the Laboratory are a 5 MW heavy-water cooled and moderated general-purpose research reactor (CP-5) fueled with fully-enriched uranium; a 200 kW light-water cooled and moderated biological research reactor (JANUS) fueled with fully-enriched uranium; one critical assembly, or zero power reactor (ZPR-9), that is fueled at various times with plutonium, uranium, or a combination of the two; the Argonne Thermal Source Reactor (ATSR), a 10 kW research reactor fueled with enriched uranium; a 12.5 GeV proton accelerator, the Zero Gradient Synchrotron (ZGS); a 60-inch cyclotron; several other charged particle accelerators; chemical and metallurgical plutonium laboratories; and several hot cells and laboratories designed for work with irradiated fuel elements and with multicurie quantities of the actinide elements.

B. Description of Site

Argonne National Laboratory (Illinois site) occupies the central 1,700 acres (6.88 sq km) of a 3,740-acre (15.14 sq km) tract in DuPage County, 27 miles (43 km) southwest of downtown Chicago, and 24 miles (39 km) due west of Lake Michigan. It lies in the Des Plaines River Valley, south of Interstate

Highway 55 and west of Illinois Highway 83. Figures 1 and 2 are maps of the site and of the surrounding area. The 2,040-acre (8.26 sq km) area surrounding the site (Waterfall Glen Forest Preserve) was formerly Argonne property, but was deeded to the DuPage County Forest Preserve District in 1973 for their use as a public recreational area, nature preserve, and demonstration forest.

The terrain is gently rolling, partially-wooded, former prairie and farmland. The grounds contain a number of ponds and small streams, the principal one being Sawmill Creek, which runs through the site in a southerly direction and enters the Des Plaines River about 1.3 miles (2.1 km) southeast of the center of the site. The land is drained primarily by Sawmill Creek, although the extreme southern portion drains directly into the Des Plaines River, which flows along the southern boundary of the Forest Preserve. This river flows southwest until it joins the Kankakee River about 30 miles (48 km) southwest of the Laboratory to form the Illinois River.

The largest topographical feature is the Des Plaines River channel, about 1 mile (1.6 km) wide. This channel contains both the River and the Chicago Sanitary and Ship Canal. Their presence extends the uninhabited area about 1 mile (1.6 km) south of the site. The elevation of the channel surface is 578 feet (176 m) above sea level. Bluffs, which comprise the south border of the site, rise from the channel at varying slope angles of 15° to 60°, reaching an average elevation of 650 feet (198 m) above sea level at the top. The land then slopes gradually upward reaching the average site elevation of 725 feet (221 m) above sea level at 3,000 feet (914 m) from the bluffs. Several large ravines oriented in a north-south direction are located in the southern portion of the site. The bluffs and ravines generally are forested with deciduous trees of an average height of 50-60 feet (15-18 m). The remaining portion of the site changes in elevation by no more than 25 feet (7.6 m) in a distance of 500 horizontal feet (152 m). In the southern portion of the Forest Preserve, the Chicago District Pipe Line Co. and the Atchison, Topeka, and Santa Fe Railroad have rights-of-way.

C. Meteorology

The climate of the area is that of the upper Mississippi Valley, as moderated by Lake Michigan. A summary of the meteorological data collected on the site from 1950 to 1964 is available.⁽²⁾ Similar data have been

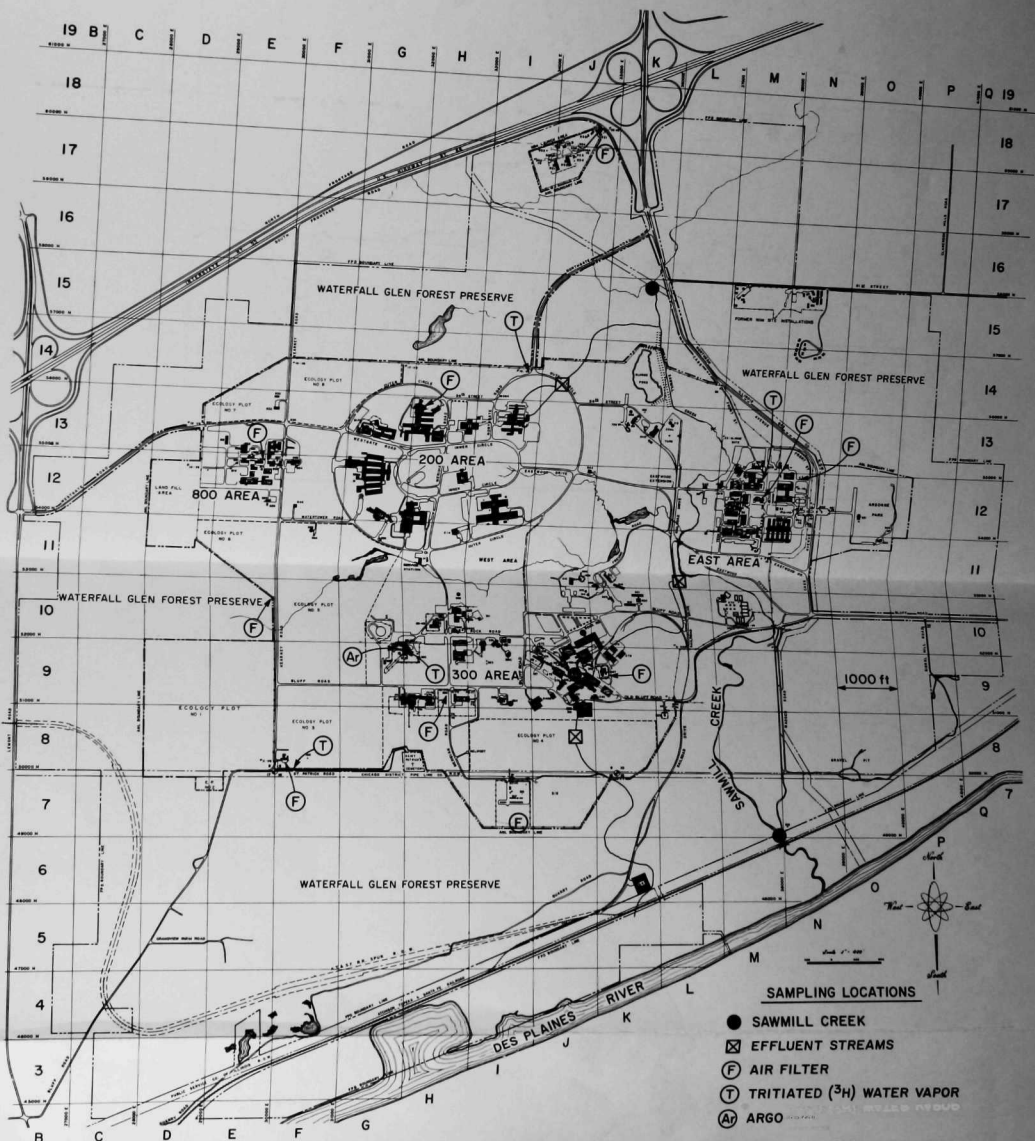


Fig. 1. Sampling Locations at Argonne National Laboratory

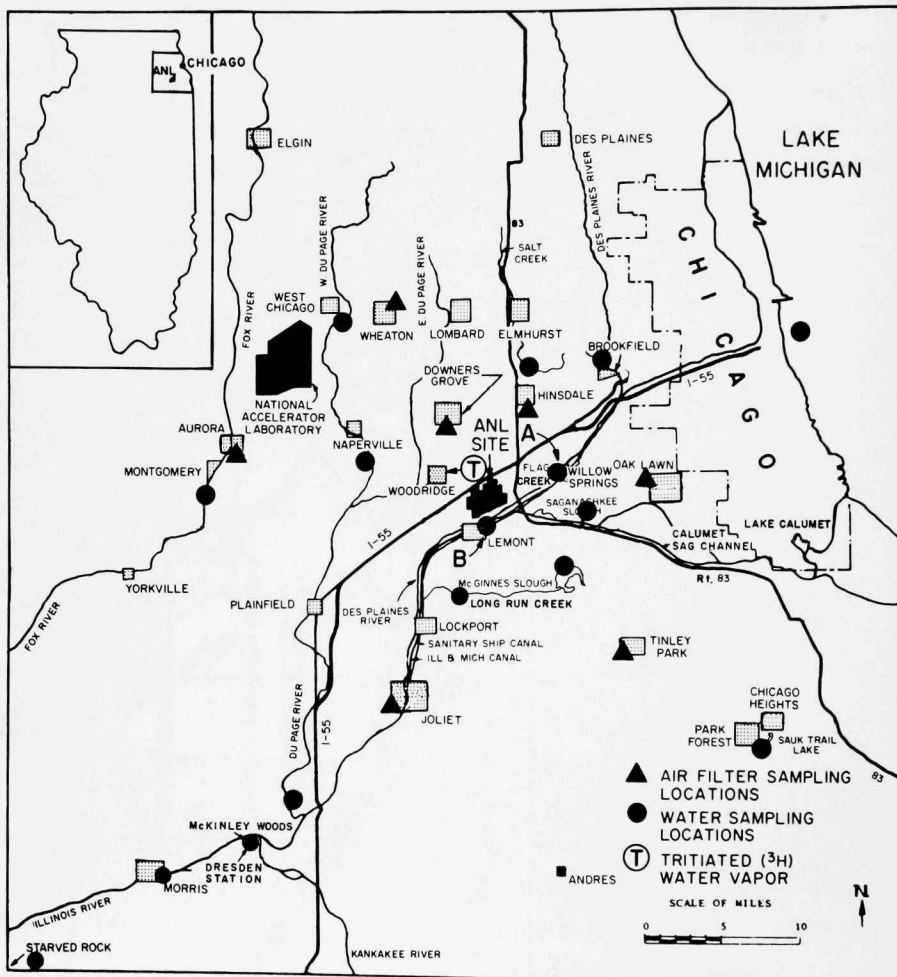


Fig. 2. Sampling Locations near Argonne National Laboratory

collected since 1964, but have not been tabulated and published. The 15-year summary, however, gives an adequate sample of the climate, and the following information is taken from this reference. The average monthly and annual wind roses are shown in Figure 3. The wind roses are essentially polar coordinate plots where the lengths of the radii represent percentage frequency of wind speeds in classes of 4 to 12 mph (6.4 to 19.3 km/hr), 4 to 24 mph (6.4 to 38.6 km/hr), and greater than 4 mph (6.4 km/hr). The direction of the radii represents the direction from which the wind blows. For example, on the 15-year summary rose for January in Figure 3 the three points plotted on the spoke extending horizontally to the left represent the wind speed distribution when the wind is blowing from the ten-degree sector, 266° to 275° . These points indicate that about 3.8 percent of the wind observations in January were from the west in the speed interval 4 to 12 mph, about 5.1 percent in the speed interval 4 to 24 mph, and about 5.2 percent in the speed interval greater than 4 mph. In the figure the curves for the latter two intervals are almost identical, and are not distinguishable on the drawing. The number 12.67 in the center of the rose represents the percent of observations of wind speed less than 4 mph in all directions.

The roses show that the predominant winds are from the southwest and west on the average, but seasonal variations are apparent. The winds are primarily westerly in January and February. In the spring, the northeast lake breeze becomes evident and continues through much of the summer. The warm southwesterly winds, brought up by the high pressure areas in the southeastern U. S., begin later in the spring, and become dominant in the fall. The winds are sufficiently variable so that monitoring for airborne releases must be carried out in all directions from the site. The daily average wind speed at 19 feet (5.8 m) above ground level varied from 5 mph (8 km/hr) in August to 10 mph (16.1 km/hr) in March. Gusts exceeded 50 mph (80 km/hr) about once a year. The mean and median wind speeds for the 15-year period were 7.6 and 7.0 mph (12.2 and 11.3 km/hr), respectively.

The air temperature data show a typical seasonal variation. Daily temperatures average from the low 20's $^{\circ}\text{F}$ (-7°C) in the winter to the 70's $^{\circ}\text{F}$ (21°C) in the summer. The extreme temperatures recorded were -20°F (-29°C) and 100°F (39°C).

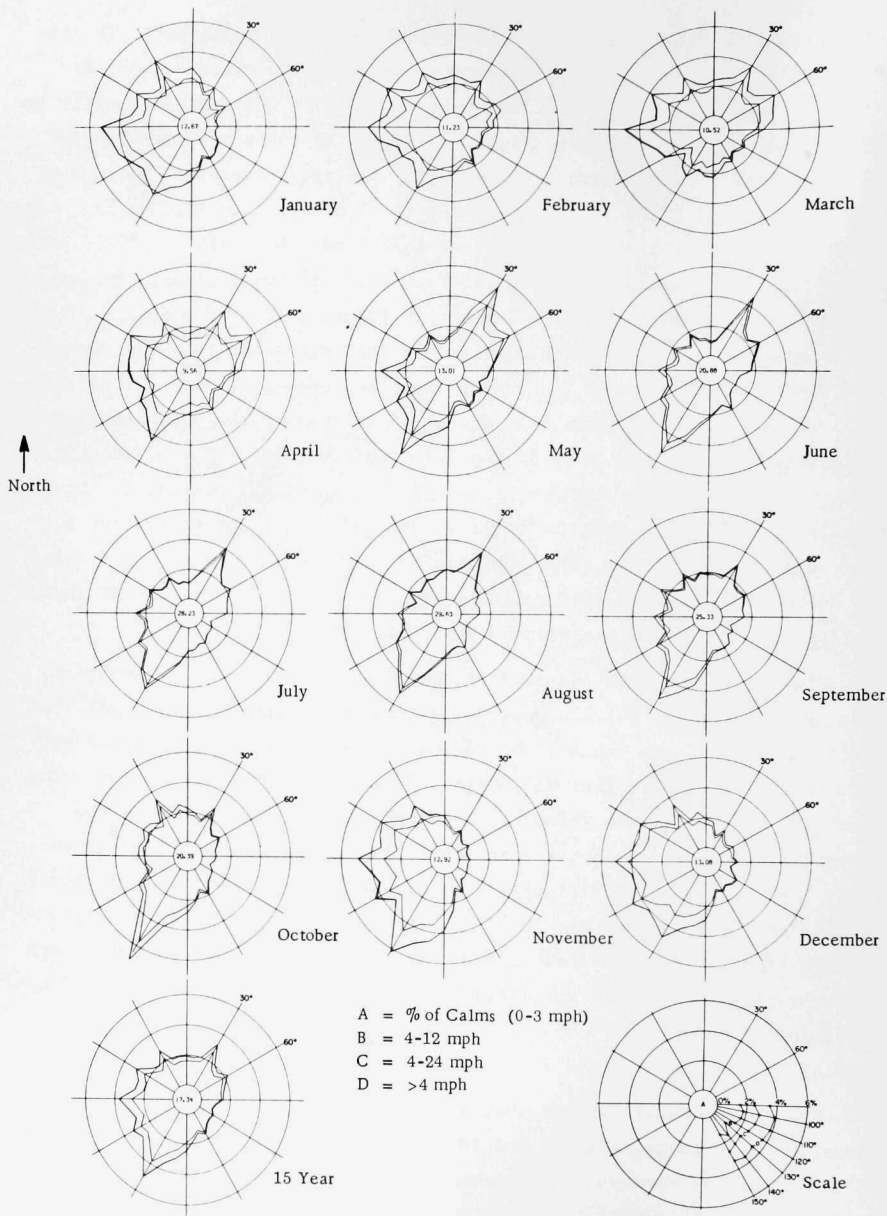


Fig. 3. Wind Roses Based on All Hourly Observations.
January 1950-December 1964, 19-foot Level.

The annual precipitation ranged between 20 and 43 inches (51 and 109 cm) and averaged 32 inches (80 cm) during the 1950-1964 period. Precipitation is heaviest in the spring and summer, when moist air is carried up from the Gulf of Mexico, and is lowest during the winter months. The monthly average varied from 1.3 inches (3.3 cm) in December to 4.3 inches (11 cm) in July.

D. Population

The area around Argonne has exhibited a large growth in population. Large areas of farmland have been converted into housing. A directional 50-mile (80 km) population distribution for the area, which is used for the population dose calculations later in this report, is shown in Table 1. The distribution, centered on the CP-5 reactor, was obtained by modifying a similar distribution provided by the Regional and Urban Studies Department at Oak Ridge National Laboratory, which was based on the 1970 U. S. census figures. The figures were adjusted in the first few miles by actual inspection of the area, and over the entire region with the use of the most recent information from the Northeastern Illinois Planning Commission (NIPC), the Northwestern Indiana Regional Planning Commission, and the individual County planning offices outside of these areas. The NIPC provided population forecasts by quarter section for 1974 in the six county area around Chicago. The Northwestern Indiana Regional Planning Commission provided the same type of data for Lake and Porter Counties. The other Illinois Counties provided their best estimates of the 1974 population in their respective areas. This count was usually a locally adjusted estimate using the State of Illinois Bureau of the Budget forecast. The 1970 census of communities and townships in the area, and a map showing their relation to the Argonne site, are given in a preceding report in the series.⁽³⁾

E. Land and Water Use

The principal stream that drains the site is Sawmill Creek. This Creek was formerly an intermittent stream, responding in flow rate largely to precipitation runoff. It now carries effluent water continuously from a municipal sewage treatment plant (Marion Brook Treatment Plant) located a few miles (kilometers) north of the site and operating at about 2 million gallons (7.6 million liters) per day. In addition, the residential development in

TABLE 1

Incremental Population Data in the Vicinity of ANL, 1974

Distance, miles	0 - 1	1 - 2	2 - 3	3 - 4	4 - 5	5-10	10-20	20-30	30-40	40-50
Distance, km	0-1.6	1.6-3.2	3.2-4.8	4.8-6.4	6.4-8.0	8-16	16-32	32-48	48-64	64-80
<u>Direction</u>										
N	0	250	2150	3100	3560	36570	188820	294520	107730	194880
NNE	0	25	1530	4530	3210	39600	336130	545820	113100	0
NE	0	160	1660	1170	1240	29400	803170	1071375	0	0
ENE	0	0	4390	755	1410	29850	737740	333865	0	0
E	0	0	130	0	10	31220	514440	252870	13720	24720
ESE	0	0	160	275	100	10760	225710	287970	295700	43080
SE	0	0	170	125	75	15230	48900	113780	21270	9140
SSE	0	20	485	440	80	1120	11820	7270	13770	18580
S	0	5	565	560	700	2910	13670	1270	26770	39320
SSW	0	15	4080	1030	730	16300	107590	8680	15130	7370
SW	0	600	150	50	100	12220	28170	4990	14460	7420
WSW	0	100	10	155	315	9140	5080	1980	5820	9990
W	0	1350	115	6480	9510	1570	47760	17720	17380	7510
WNW	0	740	35	1965	1960	21470	91000	8390	4190	50590
NW	0	180	200	2350	4995	13510	42110	74900	14910	12100
NNW	0	150	1575	1340	3650	37980	111420	146640	90780	67920
Total	0	3595	17405	24325	31645	308850	3313530	3172040	754730	492620
Cumulative Total	0	3595	21000	45325	76970	385820	3699350	6871390	7626120	8118740

the area has resulted in the collection and channeling of additional runoff water in the Creek. Treated sanitary and laboratory waste water from Argonne is discharged into Sawmill Creek at location 7M in Figure 1. In 1975, the water flow in the Creek averaged about 8.5 million gallons (32 million liters) per day upstream of the Argonne waste-water outfall; Argonne waste-water effluent averaged 0.8 million gallons (3 million liters) per day. Three streams that carry blowdown discharge water from cooling towers are considered effluents since they leave the site boundary. These are shown in Figure 1. One stream from the 200 Area leaves the site at location 14J and enters the Creek at 15K; one from the 300 Area enters Sawmill Creek at location 11L (the Creek crosses the site boundary shortly thereafter); another stream from the 300 Area crosses the site boundary at location 7K and soaks into the ground south of the site.

Sawmill Creek and the Des Plaines River above Joliet, about 13 miles (21 km) southwest of Argonne, receive very little recreational or industrial use. A few people fish in these waters downstream from Argonne, and some duck hunting takes place on the Des Plaines River. Water from the Chicago Sanitary and Ship Canal is used for some industrial purposes, such as hydro-electric generators and condensers, and for irrigation at the state prison near Joliet. It is also used as secondary cooling water by Argonne. The Canal, which carries Chicago Metropolitan Sanitary District effluent water, is used for industrial transportation and some recreational boating. Near Joliet, the River and Canal are combined into one waterway, which continues until it forms the Illinois River about 30 miles (48 km) southwest of Argonne. The Dresden Nuclear Power Station complex is located at the confluence of the Kankakee, Des Plaines, and Illinois Rivers. The plant uses water from the Kankakee for cooling, and discharges the water into the Illinois River. The first use of water for drinking is an indirect one at Peoria, on the Illinois River about 140 miles (225 km) downstream from Argonne, where River water is used to replenish ground water supplies by infiltration. In the vicinity of the Laboratory, only subsurface water (from all aquifers, shallow and deep) and Lake Michigan water is used for drinking purposes.

The principal recreational area near Argonne is Waterfall Glen Forest Preserve, which surrounds the site as described in Section I.B. and shown in Figure 1. Most of this area has received little use thus far. It was

available for hiking and skiing and its development and increased utilization by the public is expected. Sawmill Creek flows through the south portion of the Preserve on its way to the Des Plaines River. This region of the Preserve (formerly named Rocky Glen) is used principally for picnicking, hiking, and overnight camping by youth groups. During 1973, the latest year for which data is now available, approximately 7,800 individuals used the Preserve for camping and approximately 15,000 for picnics. East and southeast of Argonne and the Des Plaines River are located several large forest preserves of the Cook County Forest Preserve District. The Preserves include the two sloughs shown in Figure 2, McGinnis and Saganashkee, as well as other smaller lakes. These areas are used for picnicking, boating, fishing, and hiking. A small park located in the eastern part of the Argonne site (12-0 in Figure 1) is for the use of Argonne employees only.

The milk and principal agricultural production for 1975 in a 4,100 sq mi (10,600 sq km) area around Argonne is shown by County in Table 2. These values are estimates obtained from the Illinois Crop Reporting Service of the State Department of Agriculture.

TABLE 2

Agricultural Production Near ANL, 1975

County	Milk No. of Cows	10 ⁶ Pounds	Corn 10 ⁶ Bushels	Soybeans 10 ⁶ Bushels	Wheat 10 ³ Bushels	Oats 10 ³ Bushels
DuPage	500	5.2	2.1	0.55	198	135
Cook	600	6.2	1.7	0.76	171	122
Will	4,800	49.9	11.8	3.7	861	593
Kane	8,400	87.2	12.8	2.1	372	588
Kendall	800	8.3	9.1	1.9	250	243
Grundy	1,300	13.5	11.9	2.9	83.6	152
Lake	2,100	21.8	1.5	0.53	447	320

Note: To convert pounds into kilograms, multiply by 0.45; to convert bushels into cubic meters, multiply by 0.035.

II. SUMMARY

This is one in a series of annual reports prepared to provide the U. S. Energy Research and Development Administration (ERDA) and the public with information on the level of radioactive and chemical pollutants in the environment and on the amounts of such substances, if any, added to the environment as a result of Argonne operations. The previous report in this series is ANL-75-18. Included in this report are the results of measurements obtained in 1975 for a number of radionuclides in air, surface water, soil, grass, benthic materials, and foodstuffs; for a variety of chemical constituents in water; and for the external penetrating radiation dose.

Total alpha and beta activities, fission and activation products, plutonium, thorium, and uranium were measured continuously in air-filter samples at the site perimeter and off the site. All the off-site and perimeter samples contained only activities from natural sources and nuclear test detonations. No activity attributable to Argonne operations could be detected.

The plutonium-239,240 concentrations in air averaged, respectively, 18×10^{-18} and 17×10^{-18} $\mu\text{Ci/ml}$ * at the site perimeter and off the site, about half of the 1974 values. The monthly variations indicate a "spring maximum" in stratospheric fallout of plutonium similar to that observed in the total beta, strontium-90, and gamma-ray activities. The results indicate that the airborne plutonium was from atmospheric nuclear test detonations and there was no evidence that any of the plutonium originated at Argonne. The average plutonium-239 concentration was equivalent to 0.002% of the CG.**

Argon-41 and hydrogen-3 represent the major airborne radioactivity released from the Laboratory. The argon-41 concentration and corresponding radiation dose at the site boundary was less than the detection limit (50% of the CG) of the present sampling system, but was calculated from an atmospheric dispersion model to be 5.1×10^{-10} $\mu\text{Ci/ml}$ and 6.2 mrem/yr in the predominant north-northeast wind direction. These values are 1.2% of the CG and the

* The radioactivity units are described in Section III.

** The hazard due to a given concentration of a radioactive nuclide is assessed in this report by comparison with the Concentration Guides (CG) and annual dose limits, or Radiation Protection Standards, for uncontrolled areas specified by the U. S. Energy Research and Development Administration in Chapter 0524 of the ERDA Manual. The pertinent CGs are listed in the Appendix, Section IV.B.

non-occupational dose limit to individuals (500 mrem/yr) for uncontrolled areas. The calculated dose at 2.1 km (1.3 mi) NE, where the closest full-time residents live, is 3.5 mrem/yr. These values are consistent with penetrating radiation dose measurements made at the site perimeter. The measured hydrogen-3 concentration at the site perimeter averaged about 11×10^{-12} $\mu\text{Ci/ml}$, which is 0.006% of the CG and about eight times greater than the off-site concentration. The corresponding dose is 0.03 mrem/yr.

Argonne waste water is discharged into Sawmill Creek, and this stream was sampled above and below the site to evaluate the effect of Argonne operations on its radioactive content. The nuclides (for which analyses were made) added to the Creek in the waste water, and the percent CG of their average concentrations, were hydrogen-3, 0.45%; strontium-90, 0.2%; neptunium-237, 0.001%; plutonium-239,240, 0.0002%; americium-241, 0.0001%; and curium-244 and/or californium-249, 0.00006%. Although Sawmill Creek is not a source of potable water, the dose to an individual using water at these concentrations as his sole source of drinking water would be about 8.8 mrem/yr. Most of these nuclides are also present in fallout, and about 3.3 mrem/yr of the total dose would result from strontium-90 produced in weapons testing.

Sawmill Creek flows into the Des Plaines River, which in turn flows into the Illinois River. The radioactivity levels in the latter two streams were similar to those in other streams in the area, and the activity added to the Creek by Argonne waste water had no measurable effect on the radioactive content of either the Des Plaines or Illinois Rivers, except for one sample. This Des Plaines River sample contained hydrogen-3, attributable to Argonne operations, at a concentration of 4.7×10^{-6} $\mu\text{Ci/ml}$. If water at this concentration were consumed by an individual for an entire year, the resulting dose would be 0.8 mrem/yr, or 0.16% of the annual standard.

Plutonium concentrations in soil showed the same general range and average at the site perimeter and off the site. The average deposition of plutonium-239,240 was 2.1×10^{-3} $\mu\text{Ci/m}^2$ at the site perimeter and 2.2×10^{-3} $\mu\text{Ci/m}^2$ off the site. The corresponding plutonium-238 averages were 0.3×10^{-3} $\mu\text{Ci/m}^2$ and 0.5×10^{-3} $\mu\text{Ci/m}^2$. The plutonium content of grass was similar to that found in previous years and was about a factor of 10^4 less than soil from the same location. The results are within the range reported

by other workers for fallout from test detonations and the plutonium found in soil and grass is attributed to this source. The plutonium content of samples from beds of streams and ponds contained from 1×10^{-9} to 48×10^{-9} $\mu\text{Ci/g}$ of plutonium-239,240, a range found in previous years to be normal for fallout plutonium in such materials. The concentrations of uranium, thorium, and some gamma-ray emitters measured in soil, plant, and benthic materials were normal.

Milk from a dairy farm near the Laboratory was analyzed for several fission products, including hydrogen-3. The short-lived fission products, iodine-131 and barium-140, were not detected. Hydrogen-3 concentrations ranged from $< 200 \times 10^{-9}$ $\mu\text{Ci/ml}$ (the detection limit) to 265×10^{-9} $\mu\text{Ci/ml}$, similar to the range found in surface water. The strontium-90 and cesium-137 concentrations decreased slightly compared to 1974. The consumption of one liter of milk per day at the average concentrations would have resulted in a total dose of about 1% of the annual limit, principally from strontium-90. These radionuclides are present in milk due to fallout, and are not related to Argonne operations.

Measurements of penetrating radiation were made at several locations at the site boundary and off the site. The off-site readings averaged 94 mrem/yr, with a standard deviation of 7 mrem/yr, similar to the previous years' averages, and normal for the area. The readings at the site boundary were in the normal range except for a region along the south boundary. Here, location 7I in Figure 1, the total dose was 292 mrem/yr, about 200 mrem/yr above normal, as a result of radiation from an on-site temporary storage facility for radioactive waste. The elevated dose at 7I was 37% of the non-occupational standard for critical individuals, about one-half of the corresponding 1974 results. This location is very rarely occupied, and there were no individuals at the south boundary receiving this dose. The calculated dose rate to the residents closest to 7I, about 1 mile (1.6 km) south of the boundary, was about 0.02 mrem/yr, 0.004% of the standard for individuals in uncontrolled areas.

The levels of chemical constituents in the Argonne waste water and in Sawmill Creek were measured and results were compared to the standards adopted by the State of Illinois in 1972. Levels of mercury, hexavalent chromium, iron, and zinc in the Des Plaines River were measured to determine

whether any impact of the Argonne effluent could be seen.

The oxygen balance in the Argonne combined sanitary and laboratory waste water effluent was within acceptable levels with the exception of ammonia nitrogen in February and March. The averages during these two months exceeded the State levels by about 10%. Average concentrations of each of the other chemical constituents in this effluent were at or below the State of Illinois limits. The average level of mercury was 94% of the State limit and exceeded this value 29% of the time (i.e., in 29% of the samples). No other constituent in this effluent exceeded the State limit at any time.

Effluents from two of the three cooling tower channels used for blowdown exceeded the State limit for chromium slightly.

The average values in Sawmill Creek for oxygen balance and chemical constituents were at or below the State of Illinois limits. The average ammonia nitrogen level above the Argonne effluent was equal to the State limit, and exceeded this limit 35% of the time. The average level below the effluent was 81% of the State limit and exceeded this value in 23% of the samples. On four occasions, levels of fecal coliform in the Argonne effluents exceeded the one time limit of 400 organisms/100 ml, but at no time was the monthly limit of 200 organisms/100 ml (geometric mean) exceeded. Individual values for copper, iron, mercury, and silver exceeded State limits occasionally. Hexavalent chromium levels were in excess of the State limit 23% of the time. Samples collected in the Des Plaines River did not show any effect of Argonne effluent on levels of mercury, hexavalent chromium, iron, or zinc in the River.

The total amounts of radioactive and chemical pollutants released by Argonne to the environment were small, and did not constitute a health hazard. Any discharges from the Laboratory that exceeded acceptable standards were temporary, and when they did occur, investigations were undertaken to identify the source and reduce its discharge.

III. MONITORING RESULTS

A. Radioactivity

The radioactivity of the environment was determined by measuring the concentrations of radioactive nuclides in naturally-occurring materials and by measuring the external penetrating radiation dose. Sample collections and measurements were made principally at the site perimeter and off the site for comparison purposes. Some on-site results are also reported when they are useful in interpreting perimeter and off-site results. Since radioactivity is usually spread by air and water, the sample collection program has concentrated on these media. In addition, soil, plants, foodstuffs, precipitation, and materials from the beds of lakes and streams were also collected and analyzed.

The results of radioactivity measurements are expressed in this report in terms of microcuries per milliliter ($\mu\text{Ci/ml}$) for water, air, and milk and microcuries per gram (g) and square meter (m^2) for soil and vegetation. When a nuclide was not detected, the result is given as less than ($<$) the minimum amount detectable (detection limit) by the analytical method used. Averages, including individual results that were less than the detection limit, were calculated by one of the following two methods. If the bulk of the individual results was less than the detection limit, the average was calculated with the assumption that such results were equal to the detection limit, and the resulting average value is expressed as less than ($<$) the computed average. If only a small fraction of the individual results was less than the detection limit, the average was calculated with the assumption that such results were actually one-half of the detection limit, and the average is given as a positive value. The averages that are obtained by using these two methods under the conditions indicated are believed to give an adequate picture of the true average activity at locations where the activity not only varied greatly, but was at times not detectable.

Average values are usually accompanied by a plus-or-minus (\pm) limit value. Unless otherwise stated, this value is the 95% confidence limit calculated from the standard deviation of the average (standard error), and is a measure of the range in the concentrations encountered at that location. It does not represent the conventional error in the average of repeated

measurements on the same or identical samples. Since many of the variations observed in environmental radioactivity are not random but occur for specific reasons (e.g., nuclear testing), samples collected from the same location at different times are not replicates. The more random the variation in activity at a particular location, the closer the confidence limits will represent the actual distribution of values at that location. The averages and confidence limits should be interpreted with this in mind. When a plus-or-minus figure accompanies an individual result in this report, it represents the statistical counting error at the 95% confidence level.

The measured concentration or radiation dose is compared with appropriate standards as a means of assessing the hazard. The standards used in this report are the Concentration Guides (CGs) and annual dose limits (Radiation Protection Standards) given in ERDA Manual Chapter 0524.⁽⁴⁾ The pertinent CGs as well as the detection limits are given in the Appendix, Section IV.B. Although the CGs apply to concentrations above natural levels, the percent of CG is sometimes given in this report for activities that are primarily of natural origin for comparative purposes. Such values are enclosed in parentheses to indicate this.

1. Air

The radioactivity of particulate matter was determined by collecting and analyzing air-filter samples. The sampling locations are shown in Figures 1 and 2. Separate collections were made for radiochemical analyses and for alpha, beta, and gamma counting. The latter measurements were made on samples collected continuously on asbestos-cellulose filter paper at eight locations at the Argonne site perimeter* and at five locations off the site. Measurements are made at the perimeter because comparison between perimeter and off-site concentrations is necessary in evaluating and establishing the normal environmental concentration. If only off-site radioactivity were reported, their normality or origin could not be evaluated. Higher activities at the site perimeter may indicate radioactivity released by Argonne if the differences are greater than the error in sampling and counting. Such

* The site perimeter samplers are placed at the nearest location to the site boundary fence that provides electrical power and shelter.

results require investigation to determine the cause of the difference. The error is between 5 and 20% for most results, but approaches 100% at the detection limit.

The total alpha and beta activities in the individual weekly samples are summarized in Table 3. These measurements were made in low-background gas-flow proportional counters, and the counting efficiencies used to convert counting rates to disintegration rates were those measured for radon decay products on filter paper. The average concentrations of a number of gamma-ray emitters, as determined by gamma-ray spectrometry performed on composite weekly samples, are given in Table 4. The gamma-ray detector is a shielded 35 cm³ lithium-drifted germanium diode, calibrated for each gamma-ray emitting nuclide given in Table 4.

The alpha activities, principally due to naturally-occurring nuclides, averaged the same as 1974 and were in their normal range. About 75% of the gamma-ray activity, and a smaller fraction of the beta activity, was due to beryllium-7, principally produced in the stratosphere by cosmic-ray interactions. The remaining activity was primarily fission and activation products from nuclear test detonations. The average beta activity for the year, 7×10^{-14} $\mu\text{Ci/ml}$, was about half the 1974 average. The decrease is principally due to the decay of intermediate half-life fission products (such as zirconium-95-niobium-95, ruthenium-106-rhodium-106, and cerium-144). These fission products are the residuals of atmospheric nuclear tests by the People's Republic of China on June 26, 1973, and June 17, 1974.

The similarity of the annual averages of airborne alpha, beta, and gamma activities at the site perimeter and off the site indicates that these activities originated in a widespread source - fallout from nuclear test detonations and naturally-occurring materials - and not in a localized source such as Argonne.

Samples for radiochemical analysis were collected at locations 12N, 7I, and 9H (Figure 1) and off the site in Downers Grove (Figure 2). Collections were made on a polystyrene filter medium. The total air volume filtered for the monthly samples was about 25,000 m³. Samples were ignited at 600°C to remove organic matter and prepared for analysis by vigorous treatment with hot hydrochloric, hydrofluoric, and nitric acids. This treatment has been found in our laboratory to solubilize plutonium that had been ignited at

TABLE 3

Total Alpha and Beta Activities in Air-Filter Samples, 1975*

Month	Location	No. of Samples	Alpha Act. (10^{-15} $\mu\text{Ci/ml}$)			Beta Act. (10^{-15} $\mu\text{Ci/ml}$)		
			Av.	Min.	Max.	Av.	Min.	Max.
January	perimeter	33	1.7	0.8	2.8	76	47	111
	off-site	19	2.1	1.1	4.3	83	57	103
February	perimeter	33	1.5	0.6	3.3	96	52	130
	off-site	20	2.0	0.8	3.8	98	66	145
March	perimeter	37	1.4	0.6	3.4	133	68	208
	off-site	19	1.6	0.6	3.4	138	67	210
April	perimeter	31	2.2	0.6	5.2	150	94	250
	off-site	18	1.9	1.3	2.6	130	74	166
May	perimeter	42	2.4	0.6	6.3	106	57	172
	off-site	18	2.7	0.8	9.8	113	70	257
June	perimeter	38	2.4	0.6	5.7	58	32	106
	off-site	19	3.1	1.6	11.0	67	37	119
July	perimeter	36	2.8	0.8	6.2	60	35	166
	off-site	21	2.2	1.0	3.7	57	39	86
August	perimeter	37	2.4	1.0	4.7	37	16	79
	off-site	19	2.2	0.8	5.2	38	22	64
September	perimeter	39	2.9	1.1	8.4	29	18	47
	off-site	22	2.7	1.2	8.5	29	13	45
October	perimeter	39	3.2	1.3	7.2	32	20	44
	off-site	22	3.0	0.7	7.1	30	22	41
November	perimeter	38	2.7	1.0	7.2	31	20	48
	off-site	18	2.7	0.9	4.3	31	23	46
December	perimeter	38	2.1	0.6	4.0	34	22	48
	off-site	19	1.8	0.5	3.6	32	17	50
Annual Summary	perimeter	441	2.3 ± 0.3	0.6	8.4	70 ± 24	16	250
	off-site	234	2.3 ± 0.3	0.5	11.0	71 ± 24	13	257
Percent CG	perimeter	-	(0.023)	(0.006)	(0.084)	0.70	0.16	2.5
	off-site	-	(0.023)	(0.005)	(0.11)	0.71	0.13	2.6

*These results were obtained by measuring the samples four days after they were collected in order to avoid counting the natural radioactivity due to radon and thoron decay products. This activity is normally present in the air and disappears within four days by radioactive decay.

TABLE 4

Gamma-Ray Activity in Air-Filter Samples, 1975
(concentrations in 10^{-15} $\mu\text{Ci/ml}$)

Nuclide	Location	January	February	March	April	May	June	July	August	September	October	November	December	Annual Average	Percent CG
Antimony-125	perimeter	0.5	0.8	1.2	1.8	1.8	0.8	0.8	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	0.8 \pm 0.3	0.00009
	off-site	< 0.5	0.8	1.2	2.0	1.8	0.7	0.8	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	0.7 \pm 0.4	0.00008
Barium-140-Lanthanum-140	perimeter	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.00005
	off-site	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.00005
Beryllium-7	perimeter	59	65	93	120	120	91	115	108	81	85	86	76	92 \pm 12	(0.00023)
	off-site	54	66	88	107	118	94	110	100	78	79	80	62	86 \pm 12	(0.00022)
Cerium-141	perimeter	1.0	1.0	0.9	0.6	0.4	0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.4 \pm 0.2	0.000008
	off-site	0.9	0.9	0.8	0.6	0.5	0.2	0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.4 \pm 0.2	0.000008
Cerium-144	perimeter	9.2	13	21	28	23	11	9.2	3.8	2.0	1.6	1.2	1.1	10 \pm 5	0.005
	off-site	8.4	13	20	25	23	10	8.6	3.6	1.8	1.3	1.4	1.0	10 \pm 5	0.005
Cesium-137	perimeter	1.0	1.4	2.2	3.2	3.1	1.6	1.4	0.8	0.4	0.3	0.5	0.3	1.4 \pm 0.6	0.00028
	off-site	0.9	1.4	2.3	2.9	3.1	1.6	1.4	0.8	0.5	0.4	0.4	0.4	1.3 \pm 0.6	0.00026
Cobalt-60	perimeter	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.00003
	off-site	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.00003
Iodine-131	perimeter	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 0.005
	off-site	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 0.005
Manganese-54	perimeter	< 0.1	< 0.1	0.2	0.4	0.3	0.2	0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.2	< 0.00002
	off-site	0.1	0.2	0.2	0.2	0.4	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.2	< 0.00002
Ruthenium-103	perimeter	1.9	2.1	2.2	2.0	1.0	0.3	0.1	0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.8 \pm 0.5	0.000027
	off-site	1.6	1.8	1.9	1.8	0.8	0.3	0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.7 \pm 0.5	0.000023
Ruthenium-106-Rhodium-106	perimeter	4.6	5.6	11	15	12	6.1	5.0	2.6	1.2	0.7	0.7	0.5	5.4 \pm 2.8	0.0027
	off-site	4.4	7.1	10	13	13	5.7	4.5	2.1	1.6	0.6	0.9	0.7	5.3 \pm 2.6	0.0026
Zirconium-95-Niobium-95	perimeter	23	28	36	39	25	9.3	5.8	2.1	0.7	< 0.5	< 0.5	< 0.5	14 \pm 9	0.0014
	off-site	22	28	35	36	26	9.0	5.8	1.8	0.7	0.5	< 0.5	< 0.5	14 \pm 8	0.0014

1000°C. Plutonium and thorium were separated on an anion exchange column, electrodeposited, and their isotopic compositions determined by alpha spectrometry. Chemical recoveries were monitored by adding known amounts of plutonium-242 and thorium-234 tracers prior to ignition. Alpha spectrometry cannot distinguish between plutonium-239 and plutonium-240, and although in the remainder of the report, including the tables, only plutonium-239 is mentioned, it should be understood that the alpha activity due to the plutonium-240 isotope is also included.

The acidity of the column effluent from the anion exchange separation was adjusted to allow the extraction of uranium and of americium. The separated elements were electrodeposited and measured by alpha spectrometry. Chemical recoveries were monitored by adding known amounts of uranium-232 and americium-243 tracers prior to ignition. Following the extractions, the aqueous phase was analyzed for radiostrontium by standard radiochemical procedures. An aliquot of the original sample solution was also analyzed for uranium by a standard fluorophotometric procedure. The uranium concentrations are obtained in mass units by this technique. Since the uranium is believed to have the normal isotopic composition, the results were converted to activity units for comparison purposes by using the specific activity of natural uranium, 0.675×10^{-6} $\mu\text{Ci/g}$. The results are given in Table 5.

The average plutonium-239 concentrations decreased at all locations by about a factor of two from 1974 and are similar to those obtained by the ERDA New York Health and Safety Laboratory (HASL). Their results⁽⁵⁾ for New York City during the first eight months of the year ranged from 13×10^{-18} $\mu\text{Ci/ml}$ in August to 47×10^{-18} $\mu\text{Ci/ml}$ in April and showed the same monthly variations as those in the table. Since the monthly variations in the plutonium concentrations are the same as those of the beta and gamma-ray activities, it may be concluded that the airborne plutonium is due to fallout from nuclear tests.

The thorium and uranium concentrations in Table 5 are in the same range found during 1974 and are considered to be of natural origin. Similar uranium concentrations have been reported by other investigators.⁽⁶⁾ The percent of CG for the averages is included for completeness; the values are placed in parentheses since the concentrations are considered to be background levels. The amounts of thorium and uranium in a sample were directly related to the mass of material collected on the paper and the concentrations per

TABLE 5

Plutonium, Thorium, Uranium, and Strontium Concentrations in Air-Filter Samples, 1975
(concentrations in 10^{-18} $\mu\text{Ci}/\text{ml}$)

Month	Location ¹	Plutonium-239 ²	Plutonium-238	Thorium-232 ³	Thorium-230	Thorium-228	Uranium ³ (natural)	Strontium-90	Strontium-89
January	12N	13 \pm 1	0.3 \pm 0.3	13 \pm 1	27 \pm 2	11 \pm 1	90 \pm 5	820 \pm 80	2520 \pm 150
	9H	13 \pm 1	0.3 \pm 0.1	15 \pm 1	31 \pm 1	18 \pm 1	86 \pm 4	760 \pm 40	2820 \pm 90
February	12N	17 \pm 1	0.6 \pm 0.3	10 \pm 1	17 \pm 1	10 \pm 1	107 \pm 5	1100 \pm 90	2730 \pm 200
	9H	21 \pm 1	0.7 \pm 0.2	14 \pm 1	30 \pm 1	17 \pm 1	117 \pm 5	1500 \pm 90	4070 \pm 200
	off-site	16 \pm 2	1.2 \pm 0.9	11 \pm 2	22 \pm 4	10 \pm 2	-	940 \pm 80	2420 \pm 130
March	12N	34 \pm 2	1.7 \pm 0.6	13 \pm 1	20 \pm 1	-	60 \pm 4	1960 \pm 90	3270 \pm 200
	9H	31 \pm 3	0.4 \pm 0.1	8 \pm 1	11 \pm 2	-	40 \pm 7	3180 \pm 280	3110 \pm 580
	off-site	29 \pm 2	1.2 \pm 0.5	11 \pm 1	19 \pm 1	-	56 \pm 4	1610 \pm 40	3100 \pm 80
April	12N	43 \pm 2	2.3 \pm 0.6	16 \pm 1	22 \pm 2	-	49 \pm 3	2420 \pm 60	3510 \pm 130
	9H	38 \pm 4	0.6 \pm 1.4	4 \pm 1	7 \pm 2	-	45 \pm 8	2380 \pm 270	3010 \pm 570
	off-site	41 \pm 2	1.5 \pm 0.5	16 \pm 1	27 \pm 2	-	47 \pm 3	2160 \pm 250	3140 \pm 510
May	12N	34 \pm 2	0.4 \pm 0.3	23 \pm 1	34 \pm 2	22 \pm 1	83 \pm 7	2130 \pm 80	1510 \pm 140
	9H	30 \pm 2	1.0 \pm 0.6	8 \pm 1	17 \pm 1	8 \pm 1	56 \pm 10	1750 \pm 240	2300 \pm 570
	off-site	34 \pm 2	0.7 \pm 0.5	11 \pm 1	17 \pm 2	10 \pm 1	61 \pm 5	2010 \pm 120	1780 \pm 200
June	12N	18 \pm 1	< 0.1	17 \pm 2	30 \pm 2	15 \pm 2	72 \pm 5	1040 \pm 190	1200 \pm 380
	9H	20 \pm 2	0.4 \pm 0.6	12 \pm 2	34 \pm 3	16 \pm 2	84 \pm 7	1850 \pm 100	1420 \pm 180
	off-site	23 \pm 2	0.5 \pm 0.5	10 \pm 1	21 \pm 1	14 \pm 1	63 \pm 5	750 \pm 30	590 \pm 50
July	12N	18 \pm 1	0.1 \pm 0.3	14 \pm 1	26 \pm 1	14 \pm 1	95 \pm 6	940 \pm 40	450 \pm 60
	9H	18 \pm 3	< 0.1	11 \pm 1	19 \pm 2	10 \pm 1	169 \pm 11	1300 \pm 90	630 \pm 190
	off-site	16 \pm 1	0.2 \pm 0.4	8 \pm 1	14 \pm 1	8 \pm 1	89 \pm 6	1240 \pm 70	< 200
August	12N	10 \pm 1	< 0.1	16 \pm 1	29 \pm 2	24 \pm 2	88 \pm 6	390 \pm 90	390 \pm 360
	7I	10 \pm 1	0.4 \pm 0.4	9 \pm 1	15 \pm 2	9 \pm 1	58 \pm 6	1320 \pm 150	< 200
	off-site	7 \pm 1	< 0.1	3 \pm 1	6 \pm 1	4 \pm 1	36 \pm 3	440 \pm 140	< 200
September	12N	12 \pm 1	< 0.1	16 \pm 2	30 \pm 3	16 \pm 2	62 \pm 5	360 \pm 50	< 200
	7I	5 \pm 1	0.2 \pm 0.3	10 \pm 1	16 \pm 1	12 \pm 1	26 \pm 3	480 \pm 20	< 200
	off-site	7 \pm 1	< 0.1	4 \pm 1	7 \pm 1	4 \pm 1	19 \pm 3	350 \pm 20	< 200
October	12N	6 \pm 1	0.3 \pm 0.3	18 \pm 1	33 \pm 2	21 \pm 2	107 \pm 7	220 \pm 50	< 200
	7I	9 \pm 1	0.6 \pm 0.3	12 \pm 1	22 \pm 2	18 \pm 2	59 \pm 5	380 \pm 110	< 200
	off-site	3 \pm 1	0.3 \pm 0.2	4 \pm 1	9 \pm 1	10 \pm 1	20 \pm 3	180 \pm 40	< 200
November	12N	6 \pm 1	< 0.1	14 \pm 1	22 \pm 2	10 \pm 1	67 \pm 4	290 \pm 50	< 200
	7I	5 \pm 1	< 0.1	7 \pm 1	10 \pm 2	6 \pm 2	45 \pm 3	290 \pm 70	< 200
	off-site	5 \pm 1	< 0.1	10 \pm 1	19 \pm 2	9 \pm 1	44 \pm 3	340 \pm 30	< 200
December	12N	4 \pm 1	< 0.1	6 \pm 1	8 \pm 1	8 \pm 1	43 \pm 2	170 \pm 40	< 200
	7I	4 \pm 1	< 0.1	6 \pm 1	10 \pm 1	10 \pm 1	37 \pm 2	200 \pm 30	< 200
	off-site	5 \pm 1	< 0.1	4 \pm 1	8 \pm 1	7 \pm 1	34 \pm 2	180 \pm 50	< 200
Monthly Averages	12N	18 \pm 7	0.5 \pm 0.4	15 \pm 2	25 \pm 4	15 \pm 4	77 \pm 12	990 \pm 460	1330 \pm 770
	7I/9H	17 \pm 7	0.4 \pm 0.2	10 \pm 2	18 \pm 5	12 \pm 3	68 \pm 23	1280 \pm 530	1490 \pm 860
	off-site	17 \pm 8	0.5 \pm 0.3	8 \pm 3	15 \pm 4	8 \pm 2	47 \pm 14	930 \pm 440	1060 \pm 780
Percent CG	12N	0.002	0.00005	(0.002)	(0.008)	(0.008)	(0.002)	0.003	0.0004
	7I/9H	0.002	0.00004	(0.001)	(0.006)	(0.006)	(0.002)	0.004	0.0005
	off-site	0.002	0.00005	(0.001)	(0.005)	(0.004)	(0.002)	0.003	0.0004

¹Perimeter locations are given in terms of the grid coordinates in Figure 1.

²Plutonium-240 is included (see text).

³The concentrations in units of $\mu\text{g}/\text{m}^3$ can be obtained by multiplying the value in $\mu\text{Ci}/\text{ml}$ by 1.48×10^{12} for uranium and by 9×10^{12} for thorium-232. The amounts of the other thorium isotopes in mass units are negligible in comparison to thorium-232.

unit mass collected are quite similar to those found in soil samples. This indicates that the bulk of these elements in the air is due to resuspension of soil. Alpha spectrometric analysis of separated uranium samples for the last five months gave excellent agreement with the fluorophotometric method and established that the isotopic composition of the airborne uranium was normal. The amount of plutonium in the air samples contributed by soil, if the resuspended soil has the same plutonium concentration as the first 5 cm on the ground, ranged from 2% in April to 10% in December of the total plutonium in the samples. The results of americium-241 measurements in air for the last five months of the year were all less than the detection limit of 5×10^{-19} $\mu\text{Ci/ml}$.

The monthly variation in the concentrations of both strontium-89 and strontium-90 correlate with the results for the other fission products in Table 4. The concentrations are considered to be due to fallout, primarily from the June 26, 1973, and June 17, 1974, atmospheric nuclear tests mentioned previously, and are consistent with levels reported by HASL.⁽⁵⁾ The latter varied from 0.6×10^{-15} $\mu\text{Ci/ml}$ in January, through a maximum in April of 2.3×10^{-15} $\mu\text{Ci/ml}$, then decreased to 0.6×10^{-15} $\mu\text{Ci/ml}$ in August. There is no indication that any strontium in the air samples originated from Argonne operations. The spring increase is evident only in the plutonium and strontium activities, indicating their stratospheric-fallout origin, and not in the thorium and uranium concentrations.

Air sampling for argon-41 and hydrogen-3 (tritiated) water vapor was conducted in the exhaust stack of the CP-5 reactor (Building 330, 9H) because this reactor is the principal source of these nuclides at the Laboratory, and knowledge of their concentrations at this location serves as a source term. Tritiated water vapor measurements were also made at the site perimeter. Argon-41 was collected by filling an evacuated "Marinelli-type" container with air during reactor operation and measured by gamma-ray spectrometry. Hydrogen-3 (tritiated) water vapor in air was collected by adsorption on silica gel and was measured by counting the desorbed water in a liquid scintillation counter.

Hydrogen-3 concentrations at several locations are given in Table 6. The 12M location (1900 m east-northeast of the CP-5 reactor) may be considered a perimeter sample for CP-5. The average hydrogen-3 concentrations at all

TABLE 6

Hydrogen-3 Perimeter and Off-Site Concentrations, 1975

Month	Location *	No. of Samples	Conc. (10^{-12} $\mu\text{Ci}/\text{ml}$)			Percent CG		
			Av.	Min.	Max.	Av.	Min.	Max.
January	perimeter (14I)	8	8.8	1.0	41	0.0044	0.00050	0.020
	perimeter (12M)	2	2.6	2.6	2.6	0.0013	0.0013	0.0013
	off-site	2	0.19	0.10	0.29	0.000095	0.000050	0.00014
February	perimeter (14I)	8	23	0.53	167	0.012	0.00027	0.083
	perimeter (12M)	2	1.8	1.4	2.1	0.00090	0.00070	0.0011
	off-site	2	0.79	0.50	1.1	0.00040	0.00025	0.00055
March	perimeter (14I)	9	7.6	1.3	32	0.0038	0.00065	0.016
	perimeter (12M)	2	5.9	2.2	9.5	0.0029	0.0011	0.0048
	off-site	2	1.5	1.5	1.6	0.00075	0.00075	0.00080
April	perimeter (14I)	9	6.8	0.97	28	0.0034	0.00049	0.014
	perimeter (12M)	2	2.1	1.4	2.9	0.0011	0.00070	0.0014
	off-site	2	1.5	1.5	1.5	0.00075	0.00075	0.00075
May	perimeter (14I)	9	16	2.3	42	0.0080	0.0012	0.021
	perimeter (12M)	2	< 8.5	< 0.1	17	< 0.0042	< 0.00005	0.0085
	off-site	2	< 0.6	< 0.1	1.2	< 0.00030	< 0.00005	0.00060
June	perimeter (8F)	9	20	2.8	73	0.010	0.0014	0.036
	perimeter (14I)	9	13	0.1	33	0.0065	0.00005	0.016
	perimeter (12M)	2	11	1.1	22	0.0055	0.00055	0.011
	off-site	2	-	-	< 0.1	-	-	< 0.00005
July	perimeter (8F)	8	15	5.6	45	0.0075	0.0028	0.022
	perimeter (14I)	8	11	5.1	24	0.0055	0.0026	0.012
August	perimeter (8F)	9	21	2.2	53	0.010	0.0011	0.027
	perimeter (14I)	9	13	4.1	37	0.0065	0.0021	0.018
	perimeter (12M)	2	6.6	4.6	8.6	0.0033	0.0023	0.0043
	off-site	2	< 1.0	< 0.1	2.0	< 0.00050	< 0.00005	< 0.0010
September	perimeter (8F)	9	29	2.2	201	0.015	0.011	0.10
	perimeter (14I)	9	12	2.6	66	0.0060	0.0013	0.033
	perimeter (12M)	2	1.4	1.2	1.6	0.00070	0.00060	0.00080
	off-site	2	3.1	2.4	3.9	0.0016	0.0012	0.0020
October	perimeter (8F)	8	7.8	0.17	43	0.0039	0.000085	0.021
	perimeter (14I)	8	16	2.2	34	0.0080	0.0011	0.017
	perimeter (12M)	2	13	3.2	23	0.0065	0.0016	0.012
	off-site	2	2.0	0.25	3.7	0.0010	0.00012	0.0018
November	perimeter (8F)	9	3.0	< 0.1	13	0.0015	< 0.00005	0.0065
	perimeter (14I)	9	12	1.2	50	0.0060	0.00060	0.025
	perimeter (12M)	1	-	-	17	-	-	0.0085
December	perimeter (8F)	9	9.7	0.5	28	0.0048	0.00025	0.014
	perimeter (14I)	8	6.0	1.5	9.9	0.0030	0.00075	0.0049
	perimeter (12M)	2	6.3	1.5	11	0.0031	0.00075	0.0055
	off-site	2	0.9	0.8	1.0	0.00045	0.00040	0.00050
Annual Summary	perimeter (8F)	61	15 \pm 7	< 0.1	201	0.0075	< 0.00005	0.10
	perimeter (14I)	103	12 \pm 3	0.1	167	0.0060	0.00005	0.083
	perimeter (12M)	21	5.5 \pm 2.5	< 0.1	23	0.0028	< 0.00005	0.012
	off-site	20	1.2 \pm 0.6	< 0.1	3.9	0.00060	< 0.00005	0.0020

* Perimeter locations are given in terms of the grid coordinates in Figure 1.

perimeter locations were similar to previous years' results. The data show correlation with wind direction and indicate that, relative to the reactor, dilution to the background level occurs before reaching the site boundary in directions other than that from which the wind is blowing. The average and maximum perimeter concentrations were equivalent to 0.007% and 0.10% of the CG, respectively.

The off-site concentrations, measured about 6.2 mi (10 km) northwest of the Laboratory, were the lowest levels observed at this location in the past four years. This background level of hydrogen-3 should be subtracted from the other concentrations in Table 5 to obtain the Argonne contribution.

Argon-41 and hydrogen-3 (in the form of tritiated water) from the CP-5 reactor constitute the major portion of the gaseous radioactive effluent released from the Laboratory. During 1975, the total amount of argon-41 discharged from the reactor is estimated to be 4.5×10^4 Ci, based on a measured release rate of 1.48 Ci/MW-hr. Since the half-life of this nuclide is only 110 minutes, about 5% will decay before reaching the site boundary if the argon-41 moves with the average wind speed of 7.6 mph (12.2 km/hr). Because the half-life is short, it is appropriate and more meaningful to consider the concentration at various distances from the Laboratory in discussing argon-41 effluent discharges. By the computational method discussed in Section III.A.6, the average concentrations in the predominant wind direction (NNE) were 4.8×10^{-10} $\mu\text{Ci/ml}$ (1.1% of the CG) at 0.93 mi (1.5 km) (the site boundary); 2.2×10^{-10} $\mu\text{Ci/ml}$ (0.6% of the CG) at 1.5 mi (2.4 km); and 9×10^{-11} $\mu\text{Ci/ml}$ (0.2% of the CG) at 2.5 mi (4 km). The total amount of hydrogen-3 (as tritiated water) discharged from the CP-5 reactor was 420 Ci, based on a measured release rate of 0.048 Ci/hr. These discharges and their effect will be discussed further in Section III.A.6.a.

Other airborne effluents were considerably lower. A small amount of argon-41, about 3.2 Ci in 1975, was released from the JANUS reactor (Building 202, location 13I). In addition, the total amount of other effluents is estimated to be less than 5 Ci/yr. The principal long-lived nuclides in this group were krypton-85, estimated to be 4.2 Ci, and elemental tritium gas, estimated to be 0.7 Ci. The other nuclides, in millicurie or smaller amounts, were various fission products. The release of iodine-131 (a nuclide of particular interest) in CP-5 exhaust air was estimated to be 0.015 Ci/yr, based on

concentration measurements in the stack. The maximum concentration at 0.94 mi (1.5 km), assuming no ground deposition, would be about 1×10^{-16} $\mu\text{Ci/ml}$, or $10^{-4}\%$ of the CG.

2. Surface Water

Total (nonvolatile) alpha and beta activities were determined by counting the residue remaining after evaporation of the water, and applying counting efficiency corrections determined for uranium-233 (for alpha activity) and thallium-204 (for beta activity), respectively, to obtain disintegration rates. Hydrogen-3 was determined by liquid scintillation counting of a separate sample, and this activity does not appear in the total beta activity. Uranium was determined fluorophotometrically, and the results calculated in terms of activity with the assumption that the isotopic composition was that of natural uranium. Analyses for other radionuclides were performed by specific radiochemical separations followed by appropriate counting. One liter aliquots were used for all analyses except hydrogen-3, plutonium, neptunium, and the transplutonium nuclides. Hydrogen-3 analyses were performed by counting 10 ml in a gel system. Plutonium and neptunium analyses were performed on 10 or 50-liter samples by a plutonium chemical separation method,⁽⁷⁾ modified to include neptunium, followed by alpha spectrometry. Plutonium-236 was used to determine the plutonium yield. A modification of the plutonium procedure allowed the group separation of a fraction containing the transplutonium elements.⁽⁸⁾ Americium-243 was added to determine chemical recovery, and individual nuclides were measured by alpha spectrometry.

Argonne waste water is discharged into Sawmill Creek, a small stream that runs through the Laboratory grounds, drains surface water from much of the site, and flows into the Des Plaines River about 0.3 mile (500 meters) downstream from the waste-water outfall. Sawmill Creek was sampled upstream from the Argonne site and downstream from the waste-water outfall to determine if radioactivity was added to the stream by Argonne waste water or from surface drainage. The sampling locations are shown in Figure 1. Below the waste-water outfall, the Creek was sampled five times a week. Since it was impractical to analyze all the samples for all the nuclides and elements desired, equal portions of the samples collected each week were combined and analyzed. The results obtained in this way represent the average concentrations in the weekly samples. Above the site, samples were usually collected

once a month and were analyzed for the same radionuclides as the below-outfall samples.

Annual summaries of the results obtained for Sawmill Creek are given in Table 7. Comparison of the results, and 95% confidence limits of the averages, for the two sampling locations show that the nuclides whose presence in Creek water can be attributed to Argonne operations were hydrogen-3, strontium-90, neptunium-237, plutonium-239, americium-241, curium-244 and/or californium-249, and occasionally iodine-131, plutonium-238, and curium-242 and/or californium-252. The percentage of individual samples containing activity attributable to Argonne was 100% for hydrogen-3, neptunium-237, and plutonium-239; 80% for americium-241; 60% for curium-244 and/or californium-249; and 50% for strontium-90. The concentrations of all these nuclides were low compared to the CGs. The principal radionuclide added to the Creek by Argonne waste water, in terms of both concentration and CG, was hydrogen-3. Its average concentration was six times greater than last year, and amounted to 0.45% of the CG.

The hydrogen-3 in the Creek above the site was similar in concentration to levels found away from the Laboratory site and is characteristic of the current ambient levels in surface water. During 1975, the hydrogen-3 content of other lakes and streams ranged from $< 200 \times 10^{-9} \mu\text{Ci/ml}$ to $267 \times 10^{-9} \mu\text{Ci/ml}$ and averaged $< 205 \times 10^{-9} \mu\text{Ci/ml}$.

The average total alpha and beta activities were slightly higher above the site, indicating that at times Argonne waste water contained less of these materials than Creek water. The higher activities above the site were probably due to the water added to the Creek by a large municipal sewage treatment plant. The large amount of dissolved solids added in the sewage water is accompanied by a small amount of radioactive materials, and increases the radioactivity in the Creek water.

In addition to the natural beta activity and that added by Argonne waste water at the outfall, beta activity from nuclear detonations was detected at both sampling locations. The normal nonvolatile beta activity is approximately $8 \times 10^{-9} \mu\text{Ci/ml}$, while the contribution from the upstream municipal sewage treatment plant is another $8 \times 10^{-9} \mu\text{Ci/ml}$. It is estimated that fallout activity added about $3 \times 10^{-9} \mu\text{Ci/ml}$ to the nonvolatile beta activity at both locations and that the Argonne contribution to the water below the

TABLE 7

Radionuclides in Sawmill Creek Water, 1975

Type of Activity	Location *	No. of Samples	Concentration (10^{-9} $\mu\text{Ci/ml}$)			Av.	Percent CG	
			Av.	Min.	Max.		Min.	Max.
Alpha (nonvolatile)	16K	12	1.8 ± 0.2	1.0	2.3	(0.060)	(0.033)	(0.077)
	7M	262	1.4 ± 0.1	0.3	2.8	(0.047)	(0.010)	(0.093)
Beta (nonvolatile)	16K	12	20 ± 3	11	32	(0.067)	(0.037)	(0.11)
	7M	262	15 ± 1	8.3	30	(0.050)	(0.028)	(0.10)
Hydrogen-3	16K	12	-	-	< 200	-	-	< 0.0067
	7M	262	$(1.3 \pm 1.5) \times 10^4$	< 200	3.3×10^5	0.45	< 0.0067	11
Strontium-89	16K	12	-	-	< 2	-	-	< 0.07
	7M	262	-	-	< 2	-	-	< 0.07
Strontium-90	16K	12	< 0.58	< 0.5	0.78	< 0.19	< 0.17	0.26
	7M	262	0.66 ± 0.09	< 0.5	1.4	0.22	< 0.17	0.47
Iodine-131	16K	12	-	-	< 3	-	-	< 1
	7M	262	< 3.2	< 3	8.0	< 1.1	< 1	2.7
Barium-140	16K	12	-	-	< 2	-	-	< 0.007
	7M	262	-	-	< 2	-	-	< 0.007
Uranium (natural)**	16K	12	1.6 ± 0.2	0.9	2.1	(0.0040)	(0.0022)	(0.0052)
	7M	262	1.8 ± 0.1	1.1	3.3	(0.0045)	(0.0028)	(0.0082)
Neptunium-237	16K	12	-	-	< 0.001	-	-	< 0.00007
	7M	262	0.042 ± 0.024	0.0024	0.60	0.0014	0.00008	0.020
Plutonium-238	16K	12	-	-	< 0.002	-	-	< 0.00004
	7M	262	< 0.0026	< 0.002	0.0091	< 0.000052	< 0.00004	0.00018
Plutonium-239	16K	12	< 0.00053	< 0.0005	0.00084	< 0.000011	< 0.00001	0.000017
	7M	262	0.0094 ± 0.0026	0.0012	0.050	0.00019	0.00002	0.0010
Americium-241	16K	12	-	-	< 0.001	-	-	< 0.000025
	7M	262	0.0043 ± 0.0014	< 0.001	0.018	0.00011	< 0.000025	0.00045
Curium-242 and/or Californium-252	16K	12	-	-	< 0.001	-	-	< 0.000005
	7M	262	< 0.0012	< 0.001	0.0036	< 0.000006	< 0.000005	0.000018
Curium-244 and/or Californium-249	16K	12	-	-	< 0.001	-	-	< 0.000014
	7M	262	0.0045 ± 0.0016	< 0.001	0.025	0.000064	< 0.000014	0.00036

* Location 16K is upstream from the Argonne site and location 7M is downstream from the Argonne waste-water outfall. See Figure 1.

** Uranium concentrations in units of $\mu\text{g/l}$ can be obtained by multiplying the concentration given by 1.48×10^3 . The average concentration in the Creek then becomes $2.5 \mu\text{g/l}$.

outfall averaged about 1×10^{-9} $\mu\text{Ci/ml}$. The Argonne contribution remained the same as 1974 levels, while the fallout contribution decreased by about a factor of two. The total concentration, regardless of source, must be used in assessing the health hazard of a radionuclide not naturally present, and the percent of the CGs for all nuclides listed in Table 7 was low.

The total radioactive effluent discharged to the Creek in Argonne waste water can be estimated from the average concentrations and the volume of water carried by the Creek. These values are 170 Ci of hydrogen-3, 4.0 mCi of strontium-90 (0.33×10^{-9} $\mu\text{Ci/ml}$ is the average concentration attributed to Argonne), 0.5 mCi of neptunium-237, 0.1 mCi of plutonium-239, 0.05 mCi of americium-241, and < 0.05 mCi of curium and californium nuclides.

Samples of phytoplankton were collected at three locations in Sawmill Creek on September 4, 1975, and analyzed for several specific radionuclides. The results are given in Table 8. The location at 11L, although on-site, is considerably above the Laboratory waste-water outfall. The average concentration of plutonium-239 in the Creek water below the outfall was about 20 times that above the outfall and yet the measured plutonium-239 concentrations in the samples at all locations were similar. This would indicate that the ability of this phytoplankton to concentrate plutonium, in the form released in the waste water, is not significant and would not serve as an important component in the pathway to man.

TABLE 8

Radionuclides in Sawmill Creek Phytoplankton, 1975
(concentrations in 10^{-12} $\mu\text{Ci/g}$)

Location*	Plutonium-239	Plutonium-238	Uranium (natural)
11L	24 ± 1	< 0.1	390 ± 20
15 m below 7M Outfall	13 ± 2	0.8 ± 0.7	310 ± 20
100 m below 7M Outfall	16 ± 1	0.2 ± 0.2	620 ± 30

* Locations are given in terms of the grid coordinates in Figure 1.

The normal method of treating water samples in our laboratory is to acidify and filter each sample within an hour of collection. The filtrate is analyzed and the concentration of any radionuclide found in this water is compared to the soluble CG in ERDA Manual Chapter 0524. The soluble component is assumed to be the one that would provide the major portion of the internal dose if this water were consumed.

To measure the relative amounts of plutonium and neptunium associated with the insoluble portion of the sample, the filtered residue from several samples was also analyzed. The residue was treated in the same manner as the air-filter samples. The results are given in Table 9. The activity in the insoluble portion is expressed per volume of filtered water. Inspection of the data indicates that all but a few percent of the neptunium is in the water portion, while 50 to 80% of the plutonium is associated with the suspended solids. Thus, the plutonium is removed to a considerable extent from this water and remains suspended or may reside in the less accessible bottom material. This finding is in agreement with the general solubility properties of the most stable oxidation states of plutonium and neptunium, IV and V,

TABLE 9

Distribution of Neptunium-237 and Plutonium-239
Between Water and Residue, 1975
(concentrations in 10^{-12} $\mu\text{Ci/ml}$)

Date and Location [*]	Neptunium-237		Plutonium-239	
	Water	Residue	Water	Residue
August 25, 7M	6.7 ± 1.1	1.2 ± 0.4	5.8 ± 1.1	10.9 ± 1.2
September 2, 7M	3.3 ± 0.8	< 1	2.0 ± 0.7	2.4 ± 0.5
September 9, 7M	185 ± 9	1.6 ± 0.6	5.5 ± 1.0	21.5 ± 2.2
September 16, 7M	149 ± 7	< 1	2.9 ± 0.7	3.0 ± 0.7
November 4, 7M	183 ± 8	3.8 ± 0.9	9.4 ± 1.3	19.3 ± 2.1
November 5, 16K	< 1	< 1	< 0.5	0.6 ± 0.5

^{*} Locations are given in terms of the grid coordinates in Figure 1.

respectively, and has significant implications on environmental studies and monitoring of these two elements. Among these are that the total plutonium release cannot be judged on the basis of soluble plutonium alone, that it is important to analyze bottom material for plutonium, and that neptunium may be more important dosimetrically than plutonium at equal release rates because of its greater solubility.

Since Sawmill Creek empties into the Des Plaines River, which in turn flows into the Illinois River, the radioactivity in the latter two streams is important in assessing the contribution of Argonne waste water to the environmental radioactivity. The Des Plaines River was sampled twice a month below, and monthly above, the mouth of Sawmill Creek to determine if the radioactivity in the Creek had any effect on the activity in the River. Annual summaries of the results obtained for these two locations are given in Table 10. The average nonvolatile alpha and beta activities, 1.5×10^{-9} $\mu\text{Ci/ml}$ and 11×10^{-9} $\mu\text{Ci/ml}$, respectively, of 26 off-site surface water samples collected this year (excluding the Des Plaines River) are identical to the levels found in the Des Plaines River and is evidence that the River activity levels are normal. The activity in Sawmill Creek is usually reduced by dilution so that it is not detectable as such in the Des Plaines River. The natural nonvolatile beta activity in the River was 8×10^{-9} $\mu\text{Ci/ml}$, and the excess, 3×10^{-9} $\mu\text{Ci/ml}$, was due to fallout.

A water sample collected on October 15, 1975, at location B, contained 4.7×10^{-6} $\mu\text{Ci/ml}$ of hydrogen-3 (0.16% of the CG), while a sample collected the next day at Romeoville, Illinois, about 4.4 mi (7 km) downstream from location B, contained 5.9×10^{-7} $\mu\text{Ci/ml}$ (0.02% of the CG). The source of this tritium was ANL effluent water.

The radioactivity in samples of Illinois River water, shown in Table 11, were similar to those found in other bodies of water in the area and to the activities found previously at these same locations. Except for tritium in two Des Plaines River samples, no radioactivity originating at Argonne could be detected in the Des Plaines or Illinois Rivers.

3. Soil, Grass, and Benthic Materials

The plutonium content of soil, grass, and benthic materials was measured at the site perimeter and off the site. The object of the off-site sampling

TABLE 10

Radionuclides in Des Plaines River Water, 1975

Type of Activity	Location *	No. of Samples	Concentration (10^{-3} $\mu\text{Ci/ml}$)			Av.	Percent CG	
			Av.	Min.	Max.		Min.	Max.
Alpha (nonvolatile)	A	12	1.6 ± 0.3	0.9	2.3	(0.053)	(0.030)	(0.077)
	B	24	1.9 ± 0.3	0.6	3.6	(0.063)	(0.020)	(0.12)
Beta (nonvolatile)	A	12	11 ± 2	6.8	18	(0.37)	(0.23)	(0.60)
	B	24	12 ± 2	7.7	23	(0.40)	(0.26)	(0.77)
Hydrogen-3	A	12	< 203	< 200	230	< 0.0068	< 0.0067	0.0077
	B	24	< 400	< 200	4700	< 0.013	< 0.0067	0.16
Strontium-89	A	12	-	-	< 2	-	-	< 0.07
	B	23	-	-	< 2	-	-	< 0.07
Strontium-90	A	12	0.60 ± 0.15	< 0.5	1.1	0.20	< 0.17	0.37
	B	23	0.64 ± 0.12	< 0.5	1.1	0.21	< 0.17	0.37
Iodine-131	A	12	-	-	< 3	-	-	< 1
	B	24	-	-	< 3	-	-	< 1
Barium-140	A	12	-	-	< 2	-	-	< 0.007
	B	24	-	-	< 2	-	-	< 0.007
Uranium ** (natural)	A	12	1.4 ± 0.2	1.0	2.2	(0.0035)	(0.0025)	(0.0055)
	B	24	1.4 ± 0.2	0.8	2.7	(0.0035)	(0.0020)	(0.0068)
Neptunium-237	A	12	-	-	< 0.001	-	-	< 0.00003
	B	12	-	-	< 0.001	-	-	< 0.00003
Plutonium-238	A	12	-	-	< 0.002	-	-	< 0.00004
	B	13	-	-	< 0.002	-	-	< 0.00004
Plutonium-239	A	12	0.0005 ± 0.0002	< 0.0005	0.0014	0.000010	< 0.00001	0.000028
	B	13	0.0008 ± 0.0003	< 0.0005	0.0018	0.000016	< 0.00001	0.000036
Americium-241	A	12	-	-	< 0.001	-	-	< 0.000025
	B	11	-	-	< 0.001	-	-	< 0.000025
Curium-242 and/or Californium-252	A	12	-	-	< 0.001	-	-	< 0.000005
	B	11	-	-	< 0.001	-	-	< 0.000005
Curium-244 and/or Californium-249	A	12	-	-	< 0.001	-	-	< 0.000014
	B	11	-	-	< 0.001	-	-	< 0.000014

* Location A, near Route 45, is upstream and location B, near Lemont, is downstream from the mouth of Sawmill Creek. See Figure 2.

** Uranium concentrations in units of $\mu\text{g/l}$ can be obtained by multiplying the concentration given by 1.48×10^3 . The average concentration is $2.1 \mu\text{g/l}$.

TABLE 11

Radionuclides in Illinois River Water, 1975
(concentrations in 10^{-9} $\mu\text{Ci/ml}$)

Location	Date Collected	Alpha *	Beta *	Hydrogen-3	Uranium **	Neptunium-237	Plutonium-239
McKinley Woods State Park	June 10	1.8	12	< 200	1.3	< 0.0005	0.00019
Below Dresden Power Station	June 10	1.0	4.3	< 200	1.2	< 0.0005	0.00028
Morris	June 10	1.5	7.7	< 200	1.0	-	-
Starved Rock State Park	June 10	1.5	6.5	< 200	0.86	-	-
McKinley Woods State Park	October 16	0.48	16	267	0.81	< 0.0005	0.00017
Starved Rock State Park	October 16	1.6	9.9	< 200	1.3	-	-
Morris	October 16	0.87	11	< 200	1.1	-	-
Below Dresden Power Station	October 16	0.88	11	236	1.2	-	-

* Nonvolatile activity.

** Uranium concentrations in units of $\mu\text{g/l}$ can be obtained by multiplying the concentration by 1.48×10^9 .

was to determine the total deposition of plutonium from weapons testing for comparison with perimeter samples, and with results obtained by other organizations for samples collected at large distances from nuclear installations. This latter comparison is useful in determining if the soil activity near Argonne was normal. For this purpose, the site selection criteria and sample preparation techniques used by the ERDA New York Health and Safety Laboratory⁽⁹⁾ were used. Sites were selected in several directions and at various distances from the Laboratory. Each site was selected on the basis that it appeared, or was known to have been, undisturbed for a number of years. Attempts were made to select open, level, grassy areas that were mowed at reasonable intervals. Public parks were selected when available.

Each soil sample consisted of two cores totaling 173 cm² in area by 30 cm deep. The grass samples were obtained by collecting the grass from a 1 m² area in the immediate vicinity of a soil sample. A grab sample technique was used to obtain benthic materials. After drying, grinding, and mixing, 100 g portions of soil, benthos, and grass were analyzed by the same method described in Section III.A. for air-filter residues. Results are given in terms of oven-dried soil, benthos, or grass.

Comparison of the perimeter (Table 12) and off-site (Table 13) soil results shows that the same general range of concentrations exist in all areas for both plutonium isotopes, and it may be concluded that the plutonium in the perimeter samples resulted primarily from fallout of debris from nuclear detonations. Fallout deposition values found by other laboratories (10,11) are in the same range as those reported here, about $2-3 \times 10^{-3} \mu\text{Ci}/\text{m}^2$.

Composite monthly precipitation samples were analyzed for plutonium-239. The results are given in Table 14 along with results from 1973 and 1974 for comparison. The total 1975 deposition by precipitation was half that of 1974, a reflection of the decreased fallout from atmospheric nuclear tests, and was equivalent to 0.6% of the amount previously deposited in the soil. The increase from 1973 to 1974 can be related to announced atmospheric nuclear tests.

The concentration of plutonium-241 was determined in some plutonium fractions of 1971 soil samples by chemically separating and measuring the americium-241 which had grown-in from the plutonium-241. The concentrations of plutonium-241 in the top 5 cm of soil averaged $1 \times 10^{-7} \mu\text{Ci}/\text{g}$. The ratio,

TABLE 12

Plutonium Content of Perimeter Soil, 1975

Date Collected	Location *	Plutonium-239		Plutonium-238		$^{238}\text{Pu}/^{239}\text{Pu}$
		10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	10^{-9} $\mu\text{Ci/g}$	10^{-3} $\mu\text{Ci/m}^2$	
June 13	13N	4.7 ± 0.5	2.0 ± 0.2	0.7 ± 0.2	0.31 ± 0.07	0.15
June 13	7F	5.0 ± 0.5	2.1 ± 0.2	0.2 ± 0.1	0.07 ± 0.04	0.034
June 13	10E	4.4 ± 0.6	1.8 ± 0.2	0.5 ± 0.2	0.20 ± 0.08	0.11
June 13	8L	4.8 ± 0.7	2.0 ± 0.3	0.4 ± 0.2	0.18 ± 0.08	0.089
June 13	14H	5.3 ± 0.5	2.2 ± 0.2	0.2 ± 0.1	0.10 ± 0.04	0.046
November 6	17J	5.5 ± 0.4	2.4 ± 0.2	0.2 ± 0.1	0.08 ± 0.03	0.033
November 6	17I	5.8 ± 0.4	2.2 ± 0.2	0.4 ± 0.1	0.16 ± 0.04	0.072
November 6	18I	5.2 ± 0.4	2.3 ± 0.2	0.3 ± 0.1	0.13 ± 0.04	0.056
November 6	10M	0.9 ± 0.1	0.5 ± 0.1	0.1 ± 0.1	0.06 ± 0.03	0.14
November 7	4F	7.4 ± 0.6	3.5 ± 0.3	0.3 ± 0.1	0.12 ± 0.05	0.034
	Average	4.9 ± 1.0	2.1 ± 0.5	0.3 ± 0.1	0.14 ± 0.05	0.077

* The locations are given in terms of the grid coordinates in Figure 1.

TABLE 13

Plutonium Content of Off-Site Soil, 1975

Date Collected	Location	Plutonium-239		Plutonium-238		$^{238}\text{Pu}/^{239}\text{Pu}$
		$10^{-9} \mu\text{Ci/g}$	$10^{-3} \mu\text{Ci/m}^2$	$10^{-9} \mu\text{Ci/g}$	$10^{-3} \mu\text{Ci/m}^2$	
June 10	Channahon, IL	6.0 ± 0.4	2.3 ± 0.1	0.5 ± 0.1	0.20 ± 0.04	0.087
June 10	Dresden Lock and Dam	4.2 ± 0.4	1.9 ± 0.2	0.1 ± 0.2	0.05 ± 0.08	0.024
June 10	Romeoville, IL	5.9 ± 0.4	2.4 ± 0.2	0.2 ± 0.1	0.21 ± 0.06	0.087
June 11	Willow Springs, IL	3.3 ± 0.2	1.3 ± 0.1	0.1 ± 0.1	0.05 ± 0.02	0.040
June 12	Lemont, IL	4.4 ± 0.4	1.8 ± 0.2	0.4 ± 0.1	0.16 ± 0.05	0.091
October 16	Starved Rock State Park, IL	4.1 ± 0.3	2.0 ± 0.2	0.3 ± 0.1	0.14 ± 0.04	0.071
October 16	Morris, IL	8.6 ± 0.7	2.4 ± 0.2	0.6 ± 0.2	0.18 ± 0.04	0.074
October 22	Saganashkee Slough	8.7 ± 0.6	3.0 ± 0.2	0.8 ± 0.1	0.26 ± 0.05	0.087
October 22	McGinnis Slough	6.9 ± 0.5	2.3 ± 0.2	0.7 ± 0.1	0.23 ± 0.04	0.10
October 23	Pioneer Park, Naperville, IL	8.0 ± 0.5	2.7 ± 0.2	1.7 ± 0.2	0.56 ± 0.07	0.21
	Average	6.0 ± 1.3	2.2 ± 0.3	0.5 ± 0.3	0.20 ± 0.09	0.087

TABLE 14

Total Plutonium-239,240 Deposition
(units of 10^{-6} $\mu\text{Ci}/\text{m}^2$)

Month	1973	1974	1975
January	-	0.60 ± 0.10	1.1 ± 0.18
February	0.14 ± 0.04	1.10 ± 0.16	1.7 ± 0.19
March	0.27 ± 0.07	2.0 ± 0.22	1.5 ± 0.17
April	0.40 ± 0.07	5.2 ± 0.37	4.0 ± 0.43
May	0.50 ± 0.06	4.6 ± 0.31	1.8 ± 0.18
June	0.16 ± 0.04	3.5 ± 0.26	1.9 ± 0.18
July	0.32 ± 0.10	5.6 ± 0.37	0.48 ± 0.10
August	0.34 ± 0.14	1.0 ± 0.19	0.38 ± 0.09
September	0.27 ± 0.17	0.25 ± 0.11	0.10 ± 0.06
October	0.30 ± 0.12	0.45 ± 0.10	0.12 ± 0.08
November	0.22 ± 0.10	0.73 ± 0.12	0.08 ± 0.07
December	0.46 ± 0.12	0.71 ± 0.12	0.21 ± 0.08
Annual Average	0.31 ± 0.07	2.1 ± 1.2	1.1 ± 0.7

$^{241}\text{Pu}/^{239}\text{Pu}$ in terms of activity, in these samples ranged from 2.1 to 4.8 and averaged 3.2.

The thorium, uranium, and cesium-137 content of soil samples was also measured and the concentrations in the perimeter (Table 15) and off-site (Table 16) samples were similar. These are expected levels of the naturally-occurring thorium and uranium activities and normal fallout concentrations of cesium-137. In terms of mass, the thorium concentrations were 2.2 $\mu\text{g/g}$ and 2.5 $\mu\text{g/g}$ at the perimeter and off the site, respectively, while the uranium concentrations were 2.1 $\mu\text{g/g}$ at the perimeter and 2.4 $\mu\text{g/g}$ off-site.

The results of radioactivity measurements in grass are given in Tables 17 and 18. The grass samples were washed before analysis to remove surface soil, which contains considerably more radionuclides per gram than grass. If the grass is analyzed without washing, results are more variable and difficult to compare between locations. The perimeter and off-site plutonium concentrations are similar to each other and to results of previous years. All the results, perimeter and off-site, were within the range expected and observed from fallout. In terms of deposition, the plutonium-239 concentration was a factor of about 10^4 less than in soil from the same location. Thorium, uranium, and the detectable gamma-ray emitting nuclides were similar at the perimeter and off-site, indicating that their concentrations are due to naturally-occurring nuclides or fallout.

A study was made of the radioactivity content of the beds of streams and rivers near the perimeter and off the site. The results are given in Table 19. Plutonium results vary widely between locations and are strongly dependent on the retentiveness of the bottom material. Plutonium concentrations up to about $30 \times 10^{-9} \mu\text{Ci/g}$ may be considered normal based on previous data.⁽³⁾ The thorium and uranium concentrations were similar to soil. In terms of mass, the thorium concentrations were 1.5 $\mu\text{g/g}$ and the uranium concentrations were 2.1 $\mu\text{g/g}$. The bed of the pond at 15H needs further investigation on the basis of the results shown in Table 19. The sample contained considerable organic material, and similar material from other bodies of water have also contained elevated concentrations of radionuclides.

TABLE 15

Uranium, Thorium, and Cesium-137 in Perimeter Soil, 1975
(concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location *	Uranium (natural)	Thorium-232	Thorium-230	Thorium-228	Cesium-137
June 13	13N	2.0 \pm 0.1	0.29 \pm 0.03	0.27 \pm 0.03	0.21 \pm 0.03	0.3 \pm 0.1
June 13	7F	1.5 \pm 0.1	0.30 \pm 0.03	0.31 \pm 0.03	0.34 \pm 0.03	0.3 \pm 0.1
June 13	10E	1.9 \pm 0.1	0.16 \pm 0.04	0.25 \pm 0.05	0.30 \pm 0.05	0.3 \pm 0.1
June 13	8L	1.5 \pm 0.1	0.13 \pm 0.03	0.19 \pm 0.04	0.23 \pm 0.04	0.4 \pm 0.1
June 13	14H	1.3 \pm 0.1	-	-	-	0.3 \pm 0.1
November 6	17J	1.3 \pm 0.1	-	-	-	0.3 \pm 0.1
November 6	17I	0.86 \pm 0.05	0.31 \pm 0.03	0.36 \pm 0.04	0.31 \pm 0.03	0.3 \pm 0.1
November 6	18I	0.87 \pm 0.04	0.40 \pm 0.03	0.37 \pm 0.03	0.48 \pm 0.03	0.2 \pm 0.1
November 6	10M	1.4 \pm 0.1	0.16 \pm 0.01	0.17 \pm 0.02	0.15 \pm 0.01	< 0.1
November 7	4F	1.3 \pm 0.1	0.16 \pm 0.02	0.20 \pm 0.02	0.19 \pm 0.02	0.4 \pm 0.1
	Average	1.4 \pm 0.2	0.24 \pm 0.07	0.26 \pm 0.05	0.28 \pm 0.07	0.3 \pm 0.1

* The locations are given in terms of the grid coordinates in Figure 1.

TABLE 16

Uranium, Thorium, and Cesium-137 in Off-Site Soil, 1975
(concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location	Uranium (natural)	Thorium-232	Thorium-230	Thorium-228	Cesium-137
June 10	Channahon, IL	1.3 \pm 0.1	0.29 \pm 0.03	0.41 \pm 0.03	0.35 \pm 0.03	0.4 \pm 0.1
June 10	Dresden Lock and Dam	2.1 \pm 0.1	0.29 \pm 0.02	0.40 \pm 0.02	0.35 \pm 0.02	0.2 \pm 0.1
June 10	Romeoville, IL	1.9 \pm 0.1	0.24 \pm 0.01	0.43 \pm 0.01	0.25 \pm 0.01	0.3 \pm 0.1
June 11	Willow Springs, IL	1.5 \pm 0.1	0.23 \pm 0.03	0.25 \pm 0.03	0.20 \pm 0.03	0.3 \pm 0.1
June 12	Lemont, IL	1.7 \pm 0.1	0.25 \pm 0.03	0.30 \pm 0.03	0.21 \pm 0.03	0.2 \pm 0.1
October 16	Starved Rock State Park, IL	0.48 \pm 0.03	0.11 \pm 0.01	0.13 \pm 0.01	0.17 \pm 0.01	0.2 \pm 0.1
October 16	Morris, IL	2.4 \pm 0.1	0.18 \pm 0.04	0.70 \pm 0.09	0.21 \pm 0.05	0.4 \pm 0.1
October 22	Saganashkee Slough	2.0 \pm 0.1	0.40 \pm 0.06	0.57 \pm 0.07	0.33 \pm 0.06	0.4 \pm 0.1
October 22	McGinnis Slough	1.9 \pm 0.1	0.52 \pm 0.10	0.70 \pm 0.12	0.39 \pm 0.09	0.3 \pm 0.1
October 23	Pioneer Park Naperville, IL	0.88 \pm 0.05	0.33 \pm 0.02	0.35 \pm 0.02	0.26 \pm 0.02	0.5 \pm 0.1
	Average	1.6 \pm 0.4	0.28 \pm 0.07	0.42 \pm 0.12	0.27 \pm 0.05	0.3 \pm 0.1

TABLE 17

Plutonium Content of Grass Samples, 1975

Date Collected	Location	Plutonium-239		Plutonium-238	
		10^{-9} $\mu\text{Ci/g}$	10^{-6} $\mu\text{Ci/m}^2$	10^{-9} $\mu\text{Ci/g}$	10^{-6} $\mu\text{Ci/m}^2$
	<u>Perimeter</u> *				
June 13	13N	0.4 ± 0.2	0.13 ± 0.05	< 0.1	< 0.01
June 13	7F	1.0 ± 0.2	0.20 ± 0.04	< 0.1	< 0.01
June 13	10E	0.6 ± 0.2	0.16 ± 0.04	< 0.1	< 0.01
June 13	8L	0.2 ± 0.2	0.10 ± 0.07	< 0.1	< 0.01
June 13	14H	2.1 ± 0.5	0.49 ± 0.12	< 0.1	< 0.01
November 6	18I	0.1 ± 0.3	0.05 ± 0.11	< 0.1	< 0.01
November 6	10M	0.5 ± 0.1	0.08 ± 0.02	< 0.1	< 0.01
November 7	4F	0.4 ± 0.1	0.07 ± 0.02	< 0.1	< 0.01
	Average	0.7 ± 0.4	0.16 ± 0.10	< 0.1	< 0.01
	<u>Off-Site</u>				
June 10	Channahon, IL	0.3 ± 0.2	0.07 ± 0.06	< 0.1	< 0.01
June 10	Dresden Lock and Dam	1.0 ± 0.2	0.17 ± 0.03	< 0.1	< 0.01
June 10	Romeoville, IL	0.9 ± 0.2	0.26 ± 0.05	< 0.1	< 0.01
June 11	Willow Springs, IL	0.7 ± 0.1	0.13 ± 0.02	-	-
June 12	Lemont, IL	0.6 ± 0.1	0.12 ± 0.02	< 0.1	< 0.01
October 16	Starved Rock State Park, IL	0.5 ± 0.3	0.06 ± 0.03	< 0.1	< 0.01
October 16	Morris, IL	0.1 ± 0.4	0.02 ± 0.07	< 0.1	< 0.01
October 22	Saganashkee Slough	0.7 ± 0.2	0.12 ± 0.03	< 0.1	< 0.01
October 22	McGinnis Slough	0.7 ± 0.2	0.09 ± 0.02	< 0.1	< 0.01
October 23	Naperville, IL	< 0.1	< 0.01	< 0.1	< 0.01
	Average	0.6 ± 0.2	0.10 ± 0.05	< 0.1	< 0.01

* The perimeter locations are given in terms of the grid coordinates in Figure 1.

TABLE 18

Radionuclides in Grass Samples, 1975
(concentrations in 10^{-6} $\mu\text{Ci/g}$)

Date Collected	Location	Uranium (natural)	Thorium-232	Thorium-230	Thorium-228	Cerium-144	Cesium-137
<u>Perimeter*</u>							
June 13	13N	0.027 \pm 0.003	0.002 \pm 0.001	0.002 \pm 0.001	0.002 \pm 0.001	< 0.5	0.2 \pm 0.1
June 13	7F	0.036 \pm 0.007	0.002 \pm 0.001	0.003 \pm 0.001	0.003 \pm 0.001	0.6 \pm 0.2	< 0.1
June 13	10E	0.017 \pm 0.003	0.001 \pm 0.001	0.002 \pm 0.001	0.001 \pm 0.001	< 0.5	< 0.1
June 13	8L	0.015 \pm 0.002	0.001 \pm 0.001	0.001 \pm 0.001	0.001 \pm 0.001	< 0.5	< 0.1
June 13	14H	0.028 \pm 0.004	0.005 \pm 0.001	0.008 \pm 0.001	0.006 \pm 0.001	0.9 \pm 0.3	< 0.1
November 6	18I	0.016 \pm 0.002	0.004 \pm 0.001	0.006 \pm 0.001	0.006 \pm 0.001	< 0.5	< 0.1
November 6	10M	0.027 \pm 0.002	0.004 \pm 0.001	0.006 \pm 0.001	0.007 \pm 0.001	< 0.5	< 0.1
November 7	4F	0.031 \pm 0.002	0.006 \pm 0.001	0.009 \pm 0.001	0.005 \pm 0.001	< 0.5	< 0.1
	Average	0.025 \pm 0.006	0.003 \pm 0.001	0.005 \pm 0.002	0.004 \pm 0.002	< 0.6	< 0.2
<u>Off-Site</u>							
June 10	Channahon, IL	0.027 \pm 0.004	0.002 \pm 0.001	0.003 \pm 0.001	0.003 \pm 0.001	0.6 \pm 0.2	0.1 \pm 0.1
June 10	Dresden Lock and Dam	0.024 \pm 0.002	0.002 \pm 0.001	0.003 \pm 0.001	0.002 \pm 0.001	0.5 \pm 0.2	< 0.1
June 10	Romeoville, IL	0.022 \pm 0.002	0.001 \pm 0.001	0.002 \pm 0.001	0.002 \pm 0.001	< 0.5	< 0.1
June 11	Willow Springs, IL	0.031 \pm 0.003	0.001 \pm 0.001	0.002 \pm 0.001	0.002 \pm 0.001	< 0.5	< 0.1
June 12	Lemont, IL	0.035 \pm 0.004	0.005 \pm 0.001	0.007 \pm 0.001	0.006 \pm 0.001	< 0.5	< 0.1
October 16	Starved Rock State Park, IL	0.027 \pm 0.002	0.008 \pm 0.001	0.011 \pm 0.001	0.006 \pm 0.001	< 0.5	< 0.1
October 16	Morris, IL	0.028 \pm 0.002	0.018 \pm 0.001	0.030 \pm 0.001	0.017 \pm 0.001	< 0.5	< 0.1
October 22	Saganashkee Slough	0.022 \pm 0.001	0.005 \pm 0.001	0.008 \pm 0.001	0.005 \pm 0.001	< 0.5	< 0.1
October 22	McGinnis Slough	0.050 \pm 0.002	-	-	-	< 0.5	0.1 \pm 0.1
October 23	Naperville, IL	0.016 \pm 0.002	-	-	-	< 0.5	< 0.1
	Average	0.028 \pm 0.006	0.005 \pm 0.004	0.008 \pm 0.007	0.005 \pm 0.004	< 0.6	< 0.1

* The perimeter locations are given in terms of the grid coordinates in Figure 1.

TABLE 19

Radionuclides in Benthic Material, 1975

Date Collected	Location	Concentration (10 ⁻⁹ µCi/g)		Uranium (natural)	Concentration (10 ⁻⁶ µCi/g)			
		Plutonium-239	Plutonium-238		Thorium-232	Thorium-230	Thorium-228	Cesium-137
	<u>Perimeter</u> [*]							
September 4	100 m below 7M Outfall	14.2 ± 0.7	0.5 ± 0.1	0.94 ± 0.05	0.14 ± 0.01	0.23 ± 0.02	0.13 ± 0.01	0.1 ± 0.1
September 4	200 m below 7M Outfall	13.0 ± 0.6	0.6 ± 0.1	0.85 ± 0.04	0.20 ± 0.02	0.31 ± 0.02	0.25 ± 0.02	0.2 ± 0.1
September 4	400 m below 7M Outfall	20.9 ± 0.9	0.8 ± 0.1	0.75 ± 0.09	0.14 ± 0.01	0.22 ± 0.02	0.17 ± 0.02	0.2 ± 0.1
September 4	500 m below 7M Outfall	14.7 ± 0.6	0.7 ± 0.1	0.57 ± 0.04	0.13 ± 0.01	0.17 ± 0.02	0.14 ± 0.02	0.1 ± 0.1
September 4	16K	2.9 ± 0.3	0.1 ± 0.1	1.2 ± 0.1	0.21 ± 0.02	0.37 ± 0.02	0.21 ± 0.02	0.2 ± 0.1
November 6	15H (pond)	48.3 ± 2.9	1.3 ± 0.3	4.3 ± 0.2	0.20 ± 0.02	0.30 ± 0.02	0.18 ± 0.01	2.3 ± 0.4
	Average	19.0 ± 13.0	0.7 ± 0.3	1.4 ± 1.2	0.17 ± 0.03	0.27 ± 0.06	0.18 ± 0.04	0.5 ± 0.2
	<u>Off-Site</u>							
June 10	DuPage River, Channahon, IL	1.6 ± 0.2	0.1 ± 0.1	1.6 ± 0.1	0.25 ± 0.02	0.50 ± 0.03	0.20 ± 0.02	0.6 ± 0.2
June 11	Flagg Creek, Willow Springs, IL	9.9 ± 0.8	0.5 ± 0.2	1.2 ± 0.1	0.57 ± 0.05	0.61 ± 0.05	0.57 ± 0.05	0.3 ± 0.1
October 23	DuPage River, West Chicago, IL	0.9 ± 0.1	0.4 ± 0.1	0.71 ± 0.04	0.71 ± 0.16	0.35 ± 0.12	0.79 ± 0.18	< 0.1
	Average	4.1 ± 5.8	0.3 ± 0.2	1.2 ± 0.5	0.51 ± 0.27	0.49 ± 0.15	0.52 ± 0.34	0.3 ± 0.1

* The perimeter locations are given in terms of the grid coordinates in Figure 1. All perimeter locations were Sawmill Creek, except for the pond at 15H.

4. Foodstuffs

Raw milk was collected monthly from a local dairy farm south of Lemont and analyzed for several radioactive nuclides by methods similar to those used for water. Iodine-131 was analyzed using a batch ion-exchange method on four liters of milk and was not present in concentrations greater than its detection limit of 1×10^{-10} $\mu\text{Ci/ml}$. The cesium-137 was analyzed by an ion-exchange method followed by gamma-ray spectrometry which allowed a reduction in its detection limit to 3×10^{-9} $\mu\text{Ci/ml}$, an improvement by a factor of two over direct counting. Strontium-89 and barium-140 were not present in concentrations greater than their detection limits of 2×10^{-9} $\mu\text{Ci/ml}$. The results are given in Table 20. The average hydrogen-3 concentration in milk is identical to the concentration of hydrogen-3 found in surface water samples away from the site. The average strontium-90 and cesium-137 concentrations decreased slightly compared to 1974. These nuclides are fission products from nuclear tests and their presence in milk is not related to Argonne operations.

The concentrations given in Table 20 may be compared to the CGs for drinking water given in the Appendix, Part B. The drinking water CGs are based on an intake of 2.2 liters per day. The consumption of one liter of milk per day would result in an average intake of $< 0.003\%$ of the hydrogen-3, 1.0% of the strontium-90, and 0.009% of the cesium-137 Concentration Guides.

Several samples of garden vegetables grown 5-10 miles (8-16 km) from Argonne were obtained and the edible portion was analyzed for the same radionuclides as the grass samples. No thorium or gamma-ray emitters were detected. The plutonium and uranium results are given in Table 21, and the concentrations are expressed in terms of air-dried weight. Compared to the results for grass, the plutonium-239 concentrations are a minimum of 20 times lower, while the uranium concentrations are 5 to 25 times lower. As in the case of milk, the radioactivity is unrelated to Argonne operations, but the information is valuable as background data.

TABLE 20

Radionuclides in Milk, 1975
(concentrations in 10^{-9} $\mu\text{Ci/ml}$)

Date Collected	Hydrogen-3	Strontium-90	Cesium-137
January 8	< 200	7.4	6.3
February 5	< 200	3.7	5.0
March 5	< 200	4.2	5.5
April 2	< 200	4.6	5.0
May 7	< 200	5.1	5.4
June 4	200	10.2	3.7
July 2	-	12.2	6.0
August 6	265	9.1	3.0
September 3	< 200	6.6	< 3
October 1	< 200	8.4	3.8
November 5	< 200	1.3	< 3
December 3	< 200	3.2	< 3
Average	< 206	6.3 ± 1.9	4.0 ± 1.0

TABLE 21

Plutonium-239 and Uranium in Garden Vegetables, 1975

Food	Plutonium-239 (concentration 10^{-12} $\mu\text{Ci/g}$)	Uranium (natural) (concentration 10^{-9} $\mu\text{Ci/g}$)
Corn	3 ± 5	1.3 ± 0.2
Tomatoes	37 ± 8	1.0 ± 0.1
Cabbage	< 1	4.8 ± 0.5

5. External Penetrating Radiation

Measurements were made with calcium fluoride (dysprosium activated) thermoluminescent dosimeter chips. Each measurement was the average of three or four chips exposed in the same packet. The response of the chips was calibrated with an NBS standard radium-226 source, and the results calculated in terms of air dose. Dosimeters were exposed at a number of locations at the site boundary to determine the dose, if any, due to Argonne operations at the closest uncontrolled approaches to the Laboratory, and at several locations on the site. The latter were chosen for two purposes: to determine where abnormal doses might be encountered, and where the results might be useful in determining the origin of any abnormal dose readings obtained at the boundary. Readings were also taken at five off-site locations for comparison purposes.

The results are summarized in Tables 22 and 23, and the site boundary and on-site readings are also shown in Figure 4. Measurements were made in six successive exposure periods that varied in length from 54 to 66 days, and in total covered the period from January 3, 1975, to January 5, 1976. The results for each period were calculated in terms of annual dose for ease in comparing measurements made for different elapsed times, and were weighted according to their exposure times in calculating the annual average at each location. The error given for an average is the 95% confidence limit calculated from the standard error.

The off-site results averaged 94 mrem/yr. The standard error of the mean of all 30 results was 1.2 mrem/yr. (The standard error of the average of the five separate locations was 3 mrem/yr.) For 1972, 1973, and 1974, the

TABLE 22

Environmental Penetrating Radiation at Off-Site Locations, 1975

Location	Dose Rate (mrem/year)						Average
	Period of Measurement						
	1/3-3/10	3/10-5/9	5/9-7/2	7/2-9/3	9/3-11/5	11/5-1/5	
Downers Grove	95	90	90	97	91	93	93 ± 3
Lockport	93	93	101	95	90	89	93 ± 4
Lombard	98	92	89	96	101	102	97 ± 5
Oak Lawn	90	87	86	82	88	85	86 ± 3
Oakbrook	103	93	96	98	112	109	102 ± 8
Average	96 ± 6	91 ± 3	92 ± 7	94 ± 6	96 ± 12	96 ± 11	94 ± 7

TABLE 23

Environmental Penetrating Radiation at ANL, 1975

Location	Dose Rate (mrem/year)						Average
	1/3-3/10	3/10-5/9	Period of Measurement		9/3-11/5	11/5-1/5	
			5/9-7/2	7/2-9/3			
14L - Boundary	86	76	76	95	90	92	86 ± 8
14I - Boundary	97	91	89	105	101	108	99 ± 8
14G - Boundary	-	87	89	106	93	98	96 ± 9
13D - Boundary	73	67	67	74	75	75	74 ± 4
9/10EF - Boundary	-	96	98	102	97	100	99 ± 3
8H - Boundary	105	100	99	111	110	106	105 ± 5
8H - Center, St. Patrick's Cemetery	-	-	-	-	110	107	108 ± 9
8F - Boundary	97	-	-	-	-	-	(97)
7I - Boundary (East)	286	277	-	269	262	368	292 ± 50
7I - Boundary (West)	250	239	-	-	-	-	245 ± 24
9L - Boundary	99	87	87	91	86	88	90 ± 5
9H - 50 m SE of CP-5	3160	2740	2530	2490	2330	2320	2600 ± 320
8H - 25 m S of 316	383	459	239	759	428	296	432 ± 182
9H - 45 m E of 316	-	-	-	-	876	388	632 ± 1484
9H - 85 m E of 316	-	-	128	261	-	-	200 ± 286
9I - 45 m NE of 350, 210 m NE of 316	-	-	158	176	155	160	162 ± 13
8H - 200 m NW of Waste Storage Area (Heliport)	131	126	107	129	120	137	125 ± 10
7I - Center, Waste Storage Area	3180	3380	2510	2730	2750	4460	3180 ± 709
11J - Lodging Facilities	100	88	-	-	99	97	96 ± 8
10/11K - Lodging Facilities	-	-	-	-	81	84	82 ± 6
9J - Between ZGS Condenser and 386	112	85	83	76	101	96	93 ± 13
11G - 30 m SW of Cyclotron	110	108	107	101	95	94	102 ± 7
12F - 45 m W of 200 M-wing	83	85	84	116	-	-	92 ± 16
12M - 45 m W of 17	-	-	98	106	-	-	102 ± 19
12M - 30 m W of 55	150	145	117	136	113	105	128 ± 18
13/14H - 60 m N of Van de Graaff Accelerator	95	-	-	-	-	-	(95)
13IJ - 75 m E of 202	-	58	-	-	-	-	(58)
13J - 140 m NE of 202	-	-	93	114	107	147	116 ± 32
14/15N - Former Nike Site Frame Residence	Exterior	-	90	95	102	97	96 ± 7
	Interior	-	58	64	-	-	61 ± 13

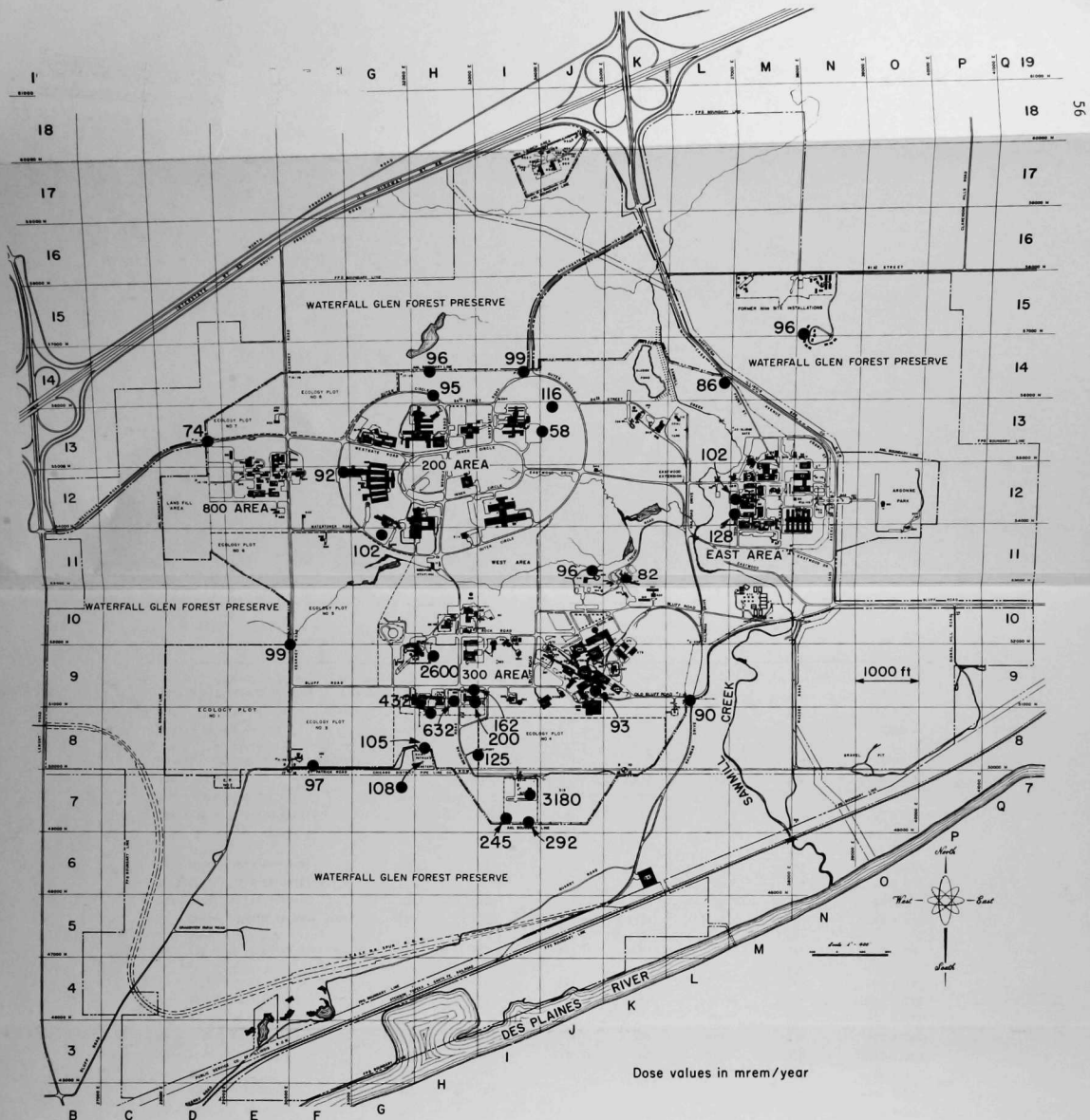


Fig. 4. Penetrating Radiation Measurements at Argonne National Laboratory

corresponding averages and standard errors were 105 ± 1.5 mrem/yr, 100 ± 2 mrem/yr, and 99 ± 2 mrem/yr, respectively. Thus, while averages for consecutive years have agreed within statistical variations at the 95% confidence level, there has been a small but steady decrease averaging about 3 mrem/yr for the past four years. The variations from year-to-year have been consistent at each location. The annual average at Downers Grove, Lockport, and Lombard have been within 4 mrem/yr of the average for all five off-site locations (usually within 2 mrem/yr). At Oak Lawn the results have been from 8 to 11 mrem/yr below, and at Oakbrook the results have been 8 to 11 mrem/yr above, the overall average. This consistency implies that the observed differences or similarities between locations are real, and that the precision of the measurements is such that comparison between perimeter and off-site readings may confidently be used to evaluate the normality of the boundary readings. Consideration must be given to the possibility that the absolute changes between years may be due to changes in the measuring system. This does not appear likely since calibration chips, exposed to the NBS standard source, are measured with each group of environmental measurements, and a new calibration curve of reader response vs dose is constructed and used each time.

The annual averages of the five off-site locations ranged from 86 ± 3 to 102 ± 8 mrem/yr. If the off-site locations are an accurate sample of the natural radiation background in the area, then annual averages at the site boundary ranging from 83 to 110 mrem/yr may be considered normal with a 95% probability, or 95% of the boundary results should have averages in this range.

To compare boundary results for individual sampling periods, the standard deviation of the 30 individual off-site results is useful. This value is 6.8 mrem/yr, so that single boundary results in the range of 94 ± 14 may be considered normal with a 95% probability, or that 95% of the measurements of a normal environmental radiation field in the area should be in the range of 80 to 108 mrem/yr.

At only one location, 7I, at the south boundary of the site, were the dose rates consistently above the normal range. This was the result of radiation from a Radioactive Waste Storage Facility in the northern half of grid 7I. Waste is packaged and temporarily kept in this area prior to

removal for permanent storage elsewhere. The net above-normal dose at this location was 150 to 200 mrem/yr, depending on the exact location measured. The location described in Table 23 as 7I-East is somewhat closer to the Storage Facility than 7I-West, and the dose rates were correspondingly higher. The excess dose rate at 7I in 1975 was similar to that found in 1974, but substantially less than in 1972 and 1973, when the average was about 320 mrem/yr above normal.

The size of the dose variations from period to period, which are at least in part statistical, and the differences between locations make it difficult to determine with high certainty when site boundary doses are only a few mrem/yr above normal and due to Argonne operations. Three criteria are used here to identify such locations: 1) the results for each sampling period are frequently above the off-site average for the same period; 2) the annual average at a location exceeds the off-site average (94 mrem/yr) plus twice the standard error in the average, 7 mrem/yr; or 3) occasional results at a particular location are significantly above the normal value for that location, although such results were still in the off-site normal range. The last criterion would apply to a location such as 13D, where the results have been consistently below the off-site average. At this location the ground contains considerable gravel, which has less radium than the usual clay subsoil, and this probably accounts for the low dose rates. Application of the first two criteria and the difficulties in interpreting small differences from natural background by TLD measurements are illustrated below.

The dose rate at the 8H boundary location is of particular interest, since it lies on the edge of St. Patrick's Cemetery, which was in use before Argonne was constructed, and which is open to visitors. The dose at this location was within the range found off-site, and thus within the expected normal distribution. However, compared to the off-site averages for each sampling period and to the annual off-site average, the 8H dose rates were about 10 mrem/yr higher. Also, two of the individual results were 2-3 mrem/yr above the expected upper limit at the 95% confidence level. Thus, a conservative upper estimate of the dose at 8H due to Argonne operations may be taken as about 10 mrem/yr. This type of comparison gave dose rates of 11 mrem/yr in 1974 and 25 mrem/yr in 1973. However, the two measurements made in the center of the Cemetery were the same as those made at the fence

line. A small decrease with distance would be expected if radiation from ANL operations contributed to the dose at the fence.

At the south end of the Laboratory site, three possible sources of external radiation exist: direct radiation from the Waste Storage Facility, direct radiation from a tandem dynamitron and low-power reactors in Buildings 314 and 316, respectively (location 9GH), and argon-41 from the CP-5 reactor at location 9H. The contribution from CP-5 to the dose at 8H is considered negligible since dose rates measured in other directions from CP-5 at the same distance, where higher doses from argon-41 would be expected because of wind direction, were less than at 8H. On the basis of other measurements made between Buildings 314-316 and the Waste Storage Facility, it is concluded that if any of the dose at the 8H boundary were due to ANL operations, the source was Buildings 314-316.

Location 14I warranted closer future examination after the 1974 measurements on the basis of the first criterion. Gamma-ray irradiation sources and the JANUS reactor in Building 202 (location 13I) are possible contributors in this area, and the measurements made at 13J, 0.087 mi (140 m) NE of Building 202 (between the 14I boundary and the building) indicate elevated dose rates for the last three sampling periods. On this basis, one may infer a contribution from Building 202 at the 14I site boundary at the rate of about 5 mrem/yr during the last six months of 1975. Most of this dose is attributed to the increased use of a cobalt-60 source during the last sampling period.

6. Potential Radiation Dose Estimates

a. Air and Water Borne Radioactivity

The radiation doses at the site boundary and off the site that could have been received by the public from radioactive materials leaving the site were calculated by two methods. Where measured radionuclide concentrations in air and water are available, the conversion of concentration to dose was based on the ratio of environmental concentrations to the Concentration Guides in ERDA Manual Chapter 0524. These Guides, and the annual radiation doses that would result from continuous exposure at these concentrations, are given in Table 24 for those nuclides whose presence in the environment is attributable to Argonne. The dose values in the table are the Radiation Protection Standards for individuals in uncontrolled areas. The second method was used

TABLE 24

Concentration-to-Dose Conversion Factors

Nuclide	Medium	Concentration [*] ($\mu\text{Ci/ml}$)	Dose [*] (rem)	Critical Organ
Americium-241	Water	4×10^{-6}	1.5	Kidney
	Water	5×10^{-6}	3	Bone
Argon-41	Air	4×10^{-8}	0.5	Whole Body
Californium-249	Water	4×10^{-6}	3	Bone
Californium-252	Water	7×10^{-6}	1.5	GI (LLI)
	Water	2×10^{-5}	3	Bone
Curium-242	Water	2×10^{-5}	1.5	GI (LLI)
	Water	2×10^{-4}	3	Bone
Curium-244	Water	7×10^{-6}	3	Bone
Hydrogen-3 (H_2O)	Air	2×10^{-7}	0.5	Whole Body
	Water	3×10^{-3}	0.5	Whole Body
Iodine-131	Air	1×10^{-10}	1.5	Thyroid
Neptunium-237	Water	3×10^{-6}	3	Bone
Plutonium-238	Water	5×10^{-6}	3	Bone
Plutonium-239	Water	5×10^{-6}	3	Bone
Strontium-90	Water	3×10^{-7}	3	Bone

* The concentrations and doses are the Radiation Protection Standards specified in ERDA Manual Chapter 0524 for individuals in uncontrolled areas.⁽⁴⁾

for argon-41, tritiated water vapor, and iodine-131 released from reactor stacks. In these cases, doses were calculated from an atmospheric dispersion model making use of a source term and meteorological data.

The principal exposure pathway for radioactive substances released from Argonne is directly from air to man by inhalation. A minor exposure route is from water to man by ingestion 140 miles (267 km) downstream from Argonne (Section I.E.). Since the dilution at this point is so great that the dose calculation is meaningless, a hypothetical calculation is made assuming Sawmill Creek water is ingested. No other exposure pathways are significant.

Argon-41 and hydrogen-3 (in the form of tritiated water) from the CP-5 reactor represent the major portion of the gaseous radioactive effluent released from the Laboratory. The concentrations and dose rates, as a function of distance from CP-5, were calculated for these two nuclides by a computer program based on an atmospheric dispersion model.⁽¹²⁾ The following parameters were used in the calculations:

- a) release rates (measured in the CP-5 exhaust stack): argon-41 1.48 Ci/MW-hr; hydrogen-3, 0.049 Ci/hr
- b) meteorological data: the 15-year average values given in ANL-7084⁽²⁾
- c) the usual parameters for stack height, building, wake, plume momentum, temperatures, etc.

The calculations were carried out to 50 miles (80 km). The argon-41 results for the first three miles (4.8 km) are given in Table 25. Doses were calculated for the mid-point of the annular interval. Thus, the dose for 0-1 mile (0-1.6 km) average is the dose at 0.5 mile (0.8 km). The highest dose rates are in the N to ENE sectors. The closest full-time residents in this area are 1.3 miles (2.1 km) from the reactor, where the dose (in the NNE direction) is 3.5 mrem/yr, less than 1% of the standard (500 mrem/yr) for individuals in uncontrolled areas. A small section of the most-used portion of the Waterfall Glen Forest Preserve, the former Rocky Glen Forest Preserve southeast of the site, begins about 0.93 mi (1,500 m) from CP-5, and in this direction the dose rate at 0.93 mi (1,500 m) is 3.2-3.8 mrem/yr.

The measurement technique for argon-41 is adequate in the vicinity of CP-5, but is not sufficiently sensitive to measure the concentration at the

TABLE 25

Argon-41 Radiation Dose From CP-5 Reactor, 1975
(millirem/year)

Sector	Distance			
	0.93 mi (1.5 km)	0-1 mi (0-1.6 km)	1-2 mi (1.6-3.2 km)	2-3 mi (3.2-4.8 km)
N	5.4	13.3	2.5	1.0
NNE	6.2	15.4	2.8	1.2
NE	4.9	12.1	2.3	0.8
ENE	5.2	13.0	2.3	1.0
E	4.1	10.3	1.8	0.8
ESE	3.9	9.6	1.8	0.7
SE	3.8	9.2	1.7	0.7
SSE	3.2	7.7	1.5	0.7
S	3.3	8.4	1.5	0.7
SSW	3.9	9.6	1.8	0.7
SW	3.3	8.2	1.5	0.7
WSW	2.8	7.1	1.3	0.5
W	2.2	5.4	1.0	0.3
WNW	2.3	5.9	1.2	0.5
NW	2.8	6.9	1.3	0.5
NNW	3.3	8.2	1.5	0.7

site boundary. However, an upper limit for the argon-41 dose at the site boundary can be estimated from the penetrating radiation dose measurements made with thermoluminescent dosimeters (TLD) and discussed in Section III.A.1. The measurements made south of the reactor are obscured by direct radiation from several sources in the vicinity, but in all other directions, including the predominant wind direction to the north-northeast, the dose rates at the site boundary were in the normal range found off-site, 94 ± 6 mrem/yr. Increases in excess of three standard deviations of the off-site average (i.e., greater than 112 mrem/yr) would have been recognized as abnormal, and on this basis the dose from argon-41 at the site boundary was less than about 15 mrem/yr. Thus, the calculated doses and those measured by TLD agree within the ability of the TLD system to detect above-normal doses.

The population data in Table 1 was used to calculate the population dose from argon-41. The results are given in Table 26, together with the average individual dose. For comparison, the table also gives the estimated natural external radiation dose, which was calculated with the assumption that the average off-site, outdoor radiation dose measured by TLD applies to the entire area within a 50-mile (80-km) radius. There are no full-time residents within 1 mile (1.6 km) of the CP-5 exhaust stack. The maximum dose to full-time residents occurs in the 1-2 mile (1.6-3.2 km) annulus in the NNE direction, where individuals would receive 5.6 mrem/yr if they were outdoors throughout the year at 1 mile (1.6 km) and 1.6 mrem/yr, outdoors at 2 miles (3.2 km).

TABLE 26

Argon-41 Average Individual and Population Dose From CP-5 Reactor, 1975

Distance (km)	Population	Dose			
		Avg. mrem/year Argon-41		man-rem/year Argon-41	
		Natural		Natural	
0-1.6	0	-	-	-	-
1.6-3.2	3,595	1.74	94	6.3	338
3.2-4.8	17,405	0.72	94	12.5	1640
0-80	8,118,740	0.020	94	162	7.63×10^5

The dose rates due to hydrogen-3 from CP-5, calculated in the same manner as for argon-41, are as follows. The maximum dose at 0.93 mi (1,500 m), in the NNE direction, is 0.013 mrem/yr. The maximum individual exposure to full-time residents in the area occurs in the 1-2 mi (1.6-3.2 km) annulus in the NNE direction, where the annual dose is calculated to be 0.008 mrem/yr. A brief summary of the results is given in Table 27. A comparison of the doses calculated from the meteorological model with the measured data from Table 6 is given in Table 28. In both cases concentrations are converted to dose as described earlier. The agreement is good in view of the large number of variables and parameters involved in obtaining both the calculated and measured values.

TABLE 27

Hydrogen-3 Average Individual and Population
Dose From CP-5 Reactor, 1975

Distance (km)	Population	Avg. mrem/year	man-rem/year
0-1.6	0	-	-
1.6-3.2	3,595	0.0040	0.014
3.2-4.8	17,405	0.0021	0.037
0-80	8,118,740	0.00017	1.38

TABLE 28

Comparison of Calculated and Measured Hydrogen-3 Dose Rates, 1975

Direction	Distance (km)	Calculated (mrem/yr)	Measured (mrem/yr)
NNE	1.5	0.013	0.027
ENE	1.9	0.0085	0.011
SW	0.45	0.038	0.034

The iodine-131 released from CP-5 would result in an individual dose of 0.002 mrem/yr at 0.93 mi (1.5 km) in the NNE sector and a population dose of about 0.001 man-rem/yr in the 1-2 mi (1.6-3.2 km) annulus. These values are so small that calculations for greater distances are not worthwhile.

The only locations where radioactivity attributable to Argonne operations could be found in off-site water were Sawmill Creek below the wastewater outfall and in the Des Plaines River, where hydrogen-3 was detected on one occasion. The concentrations of those nuclides added to Sawmill Creek by Argonne waste water, and the corresponding dose rates if water at these concentrations were used as the sole water supply by an individual are given in Table 29. In the case of strontium-90, about 50% was contributed by Argonne and the remainder by fallout, but the total concentration is included in the dose calculation since the total exposure is the figure of interest, regardless of source. For the other nuclides, essentially all of the activity may be attributed to Argonne. The dose rates were all well below the standards for individuals in uncontrolled areas. It should be emphasized that Sawmill Creek is not used for drinking or recreational purposes. There are very few fish in the stream, and they do not constitute a significant source of food for any individual.

TABLE 29

Radionuclide Concentrations and Dose Estimates
for Sawmill Creek Water, 1975

Nuclide	Conc. (avg.) 10^{-9} μ Ci/ml	Dose Rate mrem/year	Percent of Standard
Hydrogen-3	1.34×10^4	2.2	0.45
Strontium-90	0.66	6.6	0.22
Neptunium-237	0.042	0.042	0.0014
Plutonium-239	0.0094	0.0056	0.0002
Americium-241	0.0043	0.0016 (kidney) 0.0026 (bone)	0.00011 0.000087
Curium-244 and/or Californium-249	0.0045	0.0019 or 0.0034	0.00006 or 0.00011

As indicated in Table 7, occasional Creek samples (less than 10%) contained traces of plutonium-238 and curium-242 and/or californium-252, but the averages were only slightly greater than the detection limit. The annual dose due to consuming water at these concentrations can be calculated as was done for those nuclides more commonly found in Creek water, but it should be noted that the method of averaging probably exaggerates the true concentration. These annual doses are: 1.6×10^{-3} mrem/yr for plutonium-238 and from 2.6×10^{-4} to 1.8×10^{-5} mrem/yr for the other transplutonium nuclides.

One water sample from the Des Plaines River at location B contained an abnormal hydrogen-3 concentration, 4.6×10^{-6} $\mu\text{Ci/ml}$. The consumption of water at this concentration for an entire year would result in an annual dose of 0.8 mrem. The average concentration of hydrogen-3 at this location, $< 4 \times 10^{-7}$ $\mu\text{Ci/ml}$, produces a dose of < 0.067 mrem/yr. The dose from the hydrogen-3 content of 5.7×10^{-7} $\mu\text{Ci/ml}$ found at Romeoville, Illinois, in one sample would be 0.095 mrem/yr, if water at this concentration were consumed for an entire year. The average hydrogen-3 concentration of other lakes and streams found away from the Laboratory site and characteristic of the current ambient levels in surface water is $< 2.1 \times 10^{-7}$ $\mu\text{Ci/ml}$, which results in a dose of < 0.034 mrem/yr.

b. External Penetrating Radiation

The results of external penetrating radiation measurements are given in Section III.A.5. The only above-normal values unequivocally attributable to Argonne operations were measured at the south boundary adjacent to the Waste Storage Facility. At this fence line the dose from Argonne was about 175 mrem/yr. The dose at any other distance can be calculated on the basis of exponential absorption of the radiation, a decrease in intensity with the square of the distance, and an increase in intensity with distance due to the buildup factor. The closest residents to the boundary are about 1 mi (1.6 km) south, and at this distance the calculated dose rate is 0.0044 mrem/yr, if the energy of the radiation were 0.66 MeV, and 0.022 mrem/yr, if the energy were 1.3 MeV. The energy spectrum of the radiation is not known, so it is necessary to assume an energy to make the calculations. Since cesium-137 and cobalt-60 are common radionuclides, the energies of the gamma-rays from these nuclides were used in the calculations.

At the fence between the site boundary and the Cemetery (8H) the upper limit of any dose attributable to Argonne is 10 mrem/yr. An individual spending 1 hour/week at this location would then receive an annual dose of 0.06 mrem from Argonne operations. At the boundary north of Building 202 (location 14I, an unoccupied area), an Argonne-contributed dose of 2.5 mrem/yr may be inferred from the penetrating radiation measurements.

The applicable Radiation Protection Standards for whole body external radiation dose to the general population is a maximum of 500 mrem/yr to critical individuals, or, if individual doses are not known, 170 mrem/yr to a suitable sample of the exposed population. The latter criterion assumes that the maximum dose to individuals in the sample will not exceed the average by more than a factor of three.⁽¹³⁾ The doses at the south fence (7J) are about 35% of the 500 mrem/yr limit to individuals, and 103% of the 170 mrem/yr limit to the "suitable sample". However, the area south of the site boundary is heavily wooded, and the land rises steeply from the Des Plaines River. As a result, the area is relatively inaccessible and no individuals frequent this location on a regular basis. The penetrating radiation dose to individuals from Argonne operations, either those visiting the Cemetery or living south of the site, is well within all proposed radiation protection standards.

B. Chemical Pollutants

The nonradioactive environmental data contained in this report have been collected in an effort to ascertain Argonne compliance with State of Illinois regulations with regard to general use stream quality and effluent criteria as well as to verify the adequacy of Argonne's effluent pollution controls. The standards used are those adopted by the State of Illinois and approved by the Federal Government in 1972.⁽¹⁴⁾

The analytical techniques were as follows. The concentrations of barium, chromium, copper, iron, nickel, and zinc were determined using conventional atomic absorption spectrophotometry. The concentrations of silver, cadmium, and lead were determined using a flameless technique. This consists of adding the sample directly to a tantalum ribbon and drying, ashing, and atomizing the desired elements under an argon atmosphere. Mercury was determined using cold atomic absorption spectrophotometry. Fluoride levels were deter-

mined using an ion selective electrode, and the pH was determined using conventional pH electrode measurements. Dissolved oxygen, ammonia nitrogen, and hexavalent chromium levels were determined using procedures described in Standard Methods.⁽¹⁵⁾ The levels of fecal coliform were determined using the membrane filter technique.⁽¹⁵⁾ The cyanide concentration was estimated with a Hach Chemical Co. cyanide test kit. Beryllium was determined fluorophotometrically as previously described.⁽¹⁶⁾ The quality assurance studies performed are described in Appendix IV.B.

The results of the measurement of chemical constituents are expressed as milligrams per liter (mg/l) except as indicated. Averages were calculated as described in Section III.A. Yearly average values are reported with a (\pm) limit value. This value is the standard error at the 95% confidence limit and it is calculated from the standard deviation of the yearly average. Only when the sample concentrations are random does this value approach the actual distribution occurring at the sampling location. In some instances it appears that the measurements do represent a natural background value and the variation is representative of climatic conditions.

All of the results are compared to the appropriate State limits, which are listed in Table 30. Minimum detectable amounts are included for comparison. The detection limits for the atomic absorption methods represent twice the background variation, which is commonly used for this purpose. Detection limits for ion selective methods are those listed by the manufacturer, since they depend entirely on solubility considerations.

As in the past, the major emphasis has been placed on Sawmill Creek, which is a tributary of the Des Plaines River, since this is the principal route for waste water leaving the Argonne site. However, a large effort was devoted to studying cooling tower blowdown channels and a continuing emphasis was placed on the control of mercury release in the effluent.

1. Effluent Water

The major discharge of waste water from ANL operations is by way of the waste treatment plant. The water volume from this source is approximately 800,000 gallons (3×10^6 liters) per day and is comprised of about equal parts of sanitary waste water and water from Laboratory operations. The Laboratory waste water is held in 69,000 gallon (2.6×10^5 liters) tanks and

TABLE 30

Water Quality Standards and Detection Limits
(concentrations in mg/l)

Constituent	Applicable Water Type		Detection Limit
	Stream	Effluent	
Ammonia Nitrogen (as N)	1.5	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	0.1
Barium	5.0	2.0	1.0
Cadmium	0.05	0.15	0.0002
Chromium (hexavalent)	0.05	0.3	0.006
Chromium (trivalent)	1.00	1.00	0.010
Copper	0.02	1.0	0.005
Cyanide	0.025	0.025	0.020
Fluoride	1.4	2.5	0.02
Iron	1.0	2.0	0.08
Lead	0.1	0.1	0.0015
Mercury	0.0005	0.0005	0.0001
Nickel	1.0	1.0	0.2
pH	6.5-9.0	5.0-10.0	-
Silver	0.005	0.1	0.0002
Total Dissolved Solids	1000.	-	-
Zinc	1.0	1.0	0.01

is checked for radioactivity before release. The release of these tanks occurs with some periodicity at a rate of about 1,100 gallons (4,200 liters) per minute. The sanitary waste water is released at a reasonably constant rate during the entire 24-hour period. Hence, during some periods of collection, the water samples contain a higher proportion of Laboratory waste than at other times.

The performance of the sanitary waste treatment plant was monitored by the Reclamation Control Laboratory of the Plant Operations Division. This was done by analyzing twice weekly samples for biochemical oxygen demand (B.O.D.), suspended solids, and ammonia nitrogen content. Each sample was a composite of eight separate grab samples taken approximately once per hour. All analyses were performed as outlined in Standard Methods.⁽¹⁵⁾

Release of chemical pollutants from the waste treatment plant was monitored on a continuous basis during the work week. A flow proportional 24-hour sample was obtained each day and was analyzed for constituents of interest.

There are, in addition, three effluent channels from cooling tower operations which were monitored on a once-per-week schedule. These channels carry blowdown water from various cooling towers. In addition, the channel at 8J also carries some material from the treatment of Sanitary and Ship Canal water and the channel at 11L contains some photographic wastes.

The results obtained for the sanitary waste parameters are shown in Table 31. Values from January through September were obtained by analyzing only the sanitary side of the plant. For the last three months the combined sanitary and waste systems were analyzed to determine compliance with a National Pollution Discharge Elimination System (NPDES) permit obtained in 1975. The February and March averages exceeded the State of Illinois levels for ammonia nitrogen. All of the other values are within the State Guidelines.

The results obtained in the waste treatment plant effluent are shown in Table 32. All of the concentrations, with the exception of mercury in some of the samples, were within the State limits and are in the range found in the past. The levels of mercury obtained in 1975 are essentially the same as those obtained in 1974. During the past two years, a very stringent control program has been used in an effort to further reduce these levels.

TABLE 31

Performance of Sanitary Waste Treatment Plant, 1975

Month	B.O.D. ₅	Concentration (mg/l)	
		Ammonia Nitrogen	Suspended Solids
January	1.9	3.43	5.5
February	1.9	4.28	6.0
March	2.4	4.49	6.0
April	2.5	1.91	6.8
May	1.2	0.41	7.0
June	1.6	0.85	2.5
July	1.0	0.31	4.0
August	1.1	0.07	1.3
September	1.0	0.14	2.8
October	4.5	0.40	2.0
November	7.6	0.90	6.3
December	6.2	0.40	0.44
State Limit	10	2.5 (Apr.-Oct.) 4.0 (Nov.-Mar.)	15

TABLE 32

Chemical Constituents in Effluent From ANL Treatment Plant, 1975

Constituent	No. of Samples	Concentration (mg/l)			Percent of Standard (Avg.)	Percent Exceeding State Limit
		Avg.	Min.	Max.		
Barium	55	1.34 ± 0.08	0.63	1.99	67	0
Beryllium [*]	12	0.07 ± 0.021	0.03	0.12	-	-
Cadmium [*]	55	1.81 ± 0.20	0.30	3.70	1.2	0
Chromium(VI)	255	0.039 ± 0.005	0.006	0.261	1.3	0
Copper [*]	55	43.8 ± 2.8	31	82	4.4	0
Fluoride	55	0.39 ± 0.02	0.27	0.60	15.6	0
Iron	55	0.15 ± 0.02	0.06	0.45	7.5	0
Lead [*]	55	3.3 ± 1.0	0.5	27.7	3.3	0
Mercury [*]	255	0.47 ± 0.05	0.14	5.35	94	29
Nickel	55	< 0.2	-	-	20	0
pH	255	7.34	-	-	-	-
Silver [*]	55	0.83 ± 0.20	0.2	4.5	0.8	0
Zinc	55	0.24 ± 0.02	0.13	0.57	24	0

* All concentration values multiplied by 10^3 .

Although the yearly average is slightly below the State limit, approximately 30% of the samples analyzed exceeded this value. Studies are continuing to further reduce mercury concentrations.

There was a very marked decline in lead levels in 1975 as compared to previous years, when values in the range of 10-12 $\mu\text{g/l}$ were common. The 1975 average value was 3.3 $\mu\text{g/l}$. This diminution might be due, at least in part, to the more widespread use of lead-free gasoline.

Results obtained for the cooling tower effluents are shown in Table 33. The sampling site at 11L contains trivalent chromium as well as silver. The chromium(III) and silver levels result from intermittent discharges of photographic wastes from the film processing area of the ZGS Complex. Samples at this location exceeded the State limit for hexavalent chromium 23% of the time. The average concentration was 87% of the State limit. Average values for trivalent chromium and for silver at this location were well below State limits and the limits were never exceeded. The average value at 8J exceeded the State limit for hexavalent chromium by about 20%. However, one sample obtained was unusually high, 8.7 mg/l. The cause for this elevated result was located and eliminated. If this value is not included, the average value drops to 0.190 mg/l, or about 70% of the State limit. The average value for hexavalent chromium at 14J was 0.3 mg/l, which is the State limit. Approximately 90% of the values at this location exceeded the State limit.

TABLE 33

Cooling Tower Effluents, 1975

Constituent	No. of Samples	Location	Average (mg/l)	Percent of State Limit
Chromium(VI)	52	8J	0.353 ± 0.335	116
Chromium(VI)	52	11L	0.260 ± 0.094	87
Chromium(III)	52	11L	0.078 ± 0.028	7.8
Silver	52	11L	0.006 ± 0.002	6
Chromium(VI)	52	14J	0.303 ± 0.098	101

Studies are being conducted at all three effluents to reduce the hexavalent chromium levels being discharged.

2. Sawmill Creek

Samples collected for evaluation of the effect of the sanitary wastes on stream quality were obtained once per week using specially constructed sampling bottles. The sampling bottles were designed to provide temperature measurement as well as to minimize changes in oxygen content during collection.

Samples to evaluate the effect of combined sanitary and Laboratory waste on the concentrations of chemical constituents in Sawmill Creek were collected three times per week. These samples were collected 50 feet (15 m) upstream of the ANL outfall [7M (up)] and 200 feet (60 m) downstream of the outfall [7M (down)]. Additionally, samples to be examined for fecal coliform were collected five times per month using the 7M (up) location, but downstream the sample was collected immediately in front of the outfall grating to minimize contamination from the receiving stream. Once per month a sample was obtained as the water enters the site (16K), which is downstream of the Marion Brook Treatment Plant.

The data from these studies are in Tables 34 and 35. The average levels for ammonia nitrogen upstream are at the State of Illinois level and individual samples exceeded this value 35% of the time. The downstream sample averaged 81% of the State limit and exceeded this value 23% of the time. All of the samples obtained downstream that exceeded the limit could be shown to be due to upstream contamination.

The dissolved oxygen levels obtained during 1975 averaged 114% and 106% saturation for 7M (up) and 7M (down) samples. The fact that values exceeded 100% saturation is due primarily to photosynthetic activity, but the water quality must have been high to allow these levels to be reached. The fecal coliform limits require that the monthly geometric mean not exceed 200 organisms/100 ml and that no single sample exceed 400 organisms/100 ml. Samples obtained above the 7M outfall exceeded the 200 organisms/100 ml limit in May and June and there were eight instances when the 400 organisms/100 ml limit was exceeded. Samples obtained at 16K exceeded the 400 organisms/100 ml limit 60% of the time. Samples obtained at the Argonne outfall never exceeded the 200 organisms/100 ml monthly limit, but did exceed the 400 organisms/100 ml

TABLE 34

Sawmill Creek - Effect of Sanitary Waste, 1975

Constituent	Location	No. of Samples	Concentration (mg/l)			Percent of Standard	Percent Exceeding State Limit
			Av.	Min.	Max.		
Ammonia N	7M (up)	52	1.52 \pm 0.45	0.1	7.9	100	35
	7M (down)	52	1.21 \pm 0.33	0.1	5.0	81	23
Dissolved Oxygen	7M (up)	51	114%*				
	7M (down)	51	106%				
Total Dissolved Solids	7M (up)	52	1124 \pm 95	541	2028	112	64
	7M (down)	52	850 \pm 52	492	1301	85	33
<u>Organisms/100 ml</u> **							
Fecal Coliform	7M (up)	50	135			67.5	
	7M (down)	50	7			3.5	
	16K	10	184			92.0	

* Percent saturation is computed by comparing the value obtained at the temperature measured to the oxygen equilibrium value of water at that temperature.

** Average of monthly geometric mean.

TABLE 35

Chemical Constituents in Sawmill Creek, 1975

Constituent	Location *	No. of Samples	Concentration (mg/l)			Percent of Standard (Avg.)	Percent Exceeding State Level
			Avg.	Min.	Max.		
Barium	7M	50	2.13 ± 0.17	0.98	3.52	43	0
Beryllium **	7M	12	0.08 ± 0.03	0.03	0.17	-	-
Cadmium **	7M	50	1.88 ± 0.3	0.10	4.5	3.8	0
Chromium(VI)	7M (up)	52	0.016 ± 0.015	0.003	0.387	32	4
	7M (down)	150	0.034 ± 0.01	0.003	0.368	68	22
Copper **	7M	50	14.4 ± 1.6	4	37	72	13
Cyanide	7M	52	< 0.02	-	-	< 80	0
Fluoride	7M	50	0.41 ± 0.02	0.26	0.56	29	0
Iron	7M	50	0.43 ± 0.09	0.11	1.9	43	2
Lead **	7M	50	5.0 ± 1.2	1	21	5	0
Mercury **	7M (up)	52	< 0.1	< 0.1	0.1	< 20	0
	7M (down)	150	0.18 ± 0.03	0.05	1.37	36	5
Nickel	7M	50	< 0.2	-	-	< 20	0
Silver **	7M	50	1.2 ± 0.4	0.1	5.5	24	4
Zinc	7M	50	0.10 ± 0.01	0.006	0.22	10	0

* Location 7M (up) is 50 ft (15 m) upstream from the waste-water outfall. All other samples were collected 200 ft (60 m) downstream from the outfall.

** All concentration values multiplied by 10³.

limit on four occasions. These latter high levels were due to excessive chlorine demand at the treatment plant, and the cause has been located and remedial action taken. The total dissolved solids above the outfall exceeded the State limit frequently, and increased the solids content of the downstream samples.

The average results for the chemical constituents in Sawmill Creek are given in Table 35. Individual samples for hexavalent chromium at 7M (down) exceeded the State limits 22% of the time, while this limit was exceeded in 4% of the 7M samples upstream of the outfall. The latter samples contain contributions from effluents 11L and 14J, which were described earlier. Individual samples exceeded the State limit for mercury 5% of the time at the 7M (down) location. The State limit for silver was exceeded in 4% of the samples. The likely source is the 11L channel which contains photographic waste. Iron levels were exceeded in 2% of the samples. This represents soil contamination caused by erosion. As can be seen from the ANL effluent levels in Table 32, the iron source must be in the stream area since effluent levels for iron were much lower. Levels for other constituents did not exceed State limits. It is of interest to note that the level of lead is also much lower than in past years, as was observed in the effluent studies. Individual samples exceeded the limit for copper 13% of the time. It is believed that the copper is contributed largely by plumbing.

3. Des Plaines River

The effect of Sawmill Creek on the Des Plaines River was evaluated by collecting samples at Willow Springs (upstream of ANL) and at Lemont (downstream of ANL). These samples were analyzed for total mercury, hexavalent chromium, total iron, and total zinc. The results are in Table 36. In three cases, positive results were obtained for mercury in location B. In all three instances, the levels found in Sawmill Creek were lower or the same as the River results. Two of the samples had very high levels of particulates, as evidenced by iron levels of 4.3 and 4.1 mg/l. These results would indicate that the mercury is probably due to local surface run-off or to re-entrainment of mercury from the River bed.

In no case was there any indication that hexavalent chromium levels were effected by the ANL effluent. Total iron and zinc are analyzed by this

laboratory in an effort to "normalize" the water samples since both elements are naturally occurring in reasonably constant amounts.

TABLE 36

Chemical Constituents in the Des Plaines River, 1975

Constituent	Location *	No. of Samples	Concentration (mg/l)		
			Av.	Min.	Max.
Mercury (total)	B	24	< 0.1	< 0.1	0.14
Chromium (hexavalent)	B	24	< 0.006	-	< 0.006
Iron (total)	B	24	1.24	0.31	4.3
Zinc (total)	B	24	0.06	0.03	0.17
Mercury (total)	A	12	< 0.1	-	< 0.1
Chromium (Hexavalent)	A	12	< 0.006	-	< 0.006
Iron (total)	A	12	1.10	0.39	1.8
Zinc (total)	A	12	0.05	0.04	0.08

* Location A, near Route 45, is upstream and location B, near Lemont, is downstream from the mouth of Sawmill Creek. See Figure 2.

IV. APPENDIX

A. References

- (1) U. S. Energy Research and Development Administration, *Effluent and Environmental Monitoring and Reporting*, ERDA Manual Chapter 0513 and Appendix 0513 (1974).
- (2) H. Moses and M. A. Bogner, "Fifteen-Year Climatological Summary, Jan. 1, 1950-Dec. 31, 1964," *U.S.A.E.C. Report ANL-7084* (September, 1967); H. Moses and J. H. Willett, "Five-Year Climatological Summary, July 1949-June 1954," *U.S.A.E.C. Report ANL-5592*.
- (3) J. Sedlet, N. W. Golchert, and T. L. Duffy, "Environmental Monitoring at Argonne National Laboratory, Annual Report for 1973," *U.S.A.E.C. Report ANL-8078* (March, 1974).
- (4) U. S. Energy Research and Development Administration, *Standards for Radiation Protection*, ERDA Manual Chapter 0524 and Appendix 0524 (1975).
- (5) E. P. Hardy, Jr., U.S.E.R.D.A. Health and Safety Laboratory, New York, NY, Private Communication, March 26, 1976.
- (6) Environmental Protection Agency, "ERAMS Plutonium and Uranium in Air Component, January-March 1974," *Radiation Data and Reports*, 15, 719 (1974).
- (7) N. W. Golchert and J. Sedlet, "Radiochemical Determination of Plutonium in Environmental Water Samples," *Radiochem. Radioanal. Letters*, 12, 215 (1972).
- (8) M. H. Campbell (ed.), "Proceedings of the Symposium on High-Level Radioactive Waste Management," *Advances in Chemistry* (in press).
- (9) J. H. Harley (ed.), "HASL Procedure Manual," *U.S.A.E.C. Report HASL-300* (1972)
- (10) E. P. Hardy, Jr. and P. W. Krey, "Determining the Accumulated Deposit of Radionuclides by Soil Sampling and Analysis," in *Proceedings of an Environmental Plutonium Symposium*, Los Alamos Scientific Laboratory, August 4-5, 1971, *U.S.A.E.C. Report LA-4756* (December, 1971).

- (11) "Environmental Monitoring in the Vicinity of the Savannah River Plant, Annual Report for 1973," U.S.A.E.C. Report DPSPU 74-30-1 (1974).
- (12) N. A. Frigerio, K. F. Eckerman, and R. S. Stowe, "Argonne Radiological Impact Program (ARIP)," U.S.A.E.C. Report ANL/ES-26 (1973).
- (13) Federal Radiation Council, *Background Material for the Development of Radiation Protection Standards*, Report No. 1, May 13, 1960 (U. S. Government Printing Office, Washington, D. C.).
- (14) Illinois Pollution Control Board, *Rules and Regulations*, Chapter 3, 1972 (Chicago).
- (15) *Standard Methods for the Examination of Water and Waste Water*, 13th Edition, American Public Health Association, Washington, D. C., 1971.
- (16) T. L. Duffy, R. Kasper, and J. A. Wronski, "Environmental Water Studies at Argonne National Laboratory, Jan. 1968-Dec. 1969," Argonne National Laboratory Internal Report. (Available from the authors.)

B. Environmental Quality Standards, Detection Limits, Analytical Errors, and Quality Assurance

1. Standards

All of the standards and detection limits for chemical constituents, and some of those for radionuclides, are given in the main body of the report. In addition, in Table 37 are collected the air and water environmental quality standards and detection limits (minimum detectable amounts) for all radionuclides, and for those materials, for which measurements were made. These standards are the Concentration Guides given in ERDA Manual Chapter 0524, and are used in this report to assess the hazard of a measured concentration of a radioactive nuclide. Chapter 0524 distinguishes two CGs, one for occupational exposure in controlled areas and one for uncontrolled areas, beyond the site boundary where individuals can be exposed nonoccupationally, for 168 hours per week. The CGs in the table are for uncontrolled areas. For water the standard selected was for the soluble form of the radionuclide; for air the standard for the insoluble form was selected (except for iodine-131, for which the soluble form was chosen as a more conservative standard).

TABLE 37

Concentration Guides and Detection Limits
($\mu\text{Ci/ml}$)

Nuclide or Activity	Concentration Guide		Detection Limit	
	Water	Air	Water	Air
Americium-241	4×10^{-6}	-	1×10^{-12}	-
Antimony-125	-	9×10^{-10}	-	5×10^{-16}
Argon-41	-	4×10^{-8}	-	1.5×10^{-8}
Barium-140	3×10^{-5}	1×10^{-9}	2×10^{-9}	5×10^{-16}
Beryllium-7	-	4×10^{-8}	-	5×10^{-15}
Californium-249	4×10^{-6}	-	1×10^{-12}	-
Californium-252	2×10^{-5}	-	1×10^{-12}	-
Cerium-141	-	5×10^{-9}	-	5×10^{-16}
Cerium-144	-	2×10^{-10}	-	1×10^{-15}
Cesium-137	2×10^{-5}	5×10^{-10}	-	5×10^{-16}
Cobalt-60	-	3×10^{-10}	-	1×10^{-16}
Curium-242	2×10^{-5}	-	1×10^{-12}	-
Curium-244	7×10^{-6}	-	1×10^{-12}	-
Hydrogen-3	3×10^{-3}	2×10^{-7}	2×10^{-7}	1×10^{-13}
Iodine-131	3×10^{-7}	1×10^{-10}	3×10^{-9}	5×10^{-15}
Manganese-54	-	1×10^{-9}	-	5×10^{-16}
Neptunium-237	3×10^{-6}	-	1×10^{-12}	-
Plutonium-238	5×10^{-6}	1×10^{-12}	2×10^{-12}	1×10^{-19}
Plutonium-239	5×10^{-6}	1×10^{-12}	5×10^{-13}	1×10^{-19}
Ruthenium-103	-	3×10^{-9}	-	5×10^{-16}
Ruthenium-106	-	2×10^{-10}	-	1×10^{-15}
Strontium-89	3×10^{-6}	3×10^{-10}	2×10^{-9}	2×10^{-16}
Strontium-90	3×10^{-7}	3×10^{-11}	5×10^{-10}	1×10^{-17}
Thorium-228	-	2×10^{-13}	-	1×10^{-18}
Thorium-230	-	3×10^{-13}	-	1×10^{-18}
Thorium-232	-	1×10^{-12}	-	1×10^{-18}
Uranium - natural	4×10^{-5}	4×10^{-12}	2×10^{-10}	2×10^{-17}
Zirconium-95	-	1×10^{-9}	-	5×10^{-16}
Alpha *	3×10^{-6}	1×10^{-10}	2×10^{-10}	2×10^{-16}
Beta *	to 1×10^{-7}	to 1×10^{-13}	1×10^{-9}	5×10^{-16}

* The Concentration Guides for unknown mixtures depend, within the range given, on whether certain radionuclides are known to be present in concentrations less than 0.1 of their CGs, and the sum of the fraction of the CGs for all such nuclides is less than 0.25. For most total alpha and beta results given in this report, the largest CG value is applicable.

2. Detection Limits

The detection limits were chosen so that the error at the 95% confidence level is equal to the detection limit. The error in a result decreases with increasing concentration. At twice the detection limit, the error is about 50% of the measured value, and at 10 times the detection limit, the error is about 10%.

3. Quality Assurance

a. Radiochemical Analysis and Radioactivity Measurements

All nuclear instrumentation is calibrated with standard sources that are traceable to the U. S. National Bureau of Standards (NBS), if possible. If NBS standards are not available for particular nuclides, standards from the Amersham-Searle Co. are used. The equipment is usually checked on a daily basis with secondary counting standards to insure proper operation. Samples are periodically run in duplicate or with the addition of known amounts of a radionuclide to check precision and accuracy. In addition, standard and intercomparison samples distributed by the Quality Assurance Branch of the U. S. Environmental Protection Agency at Las Vegas and the International Atomic Energy Agency are analyzed regularly.

b. Chemical Analysis

With each set of atomic absorption analyses, standard amounts of trace metals were analyzed in blank solutions at concentrations corresponding to 50 and 100% of the current State limits. Recoveries were determined by comparing these results to results obtained by analyzing stream and effluent samples to which identical concentrations were added. Average recoveries ranged from about 85-100%, with a standard deviation of about 10%.

Recovery studies were also performed for ammonia nitrogen, fluoride, and hexavalent chromium analyses, and similar results were obtained.

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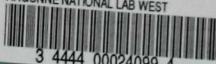
D. Acknowledgements

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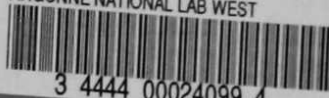
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